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Critical Pathways of Radionuclides to Man from Agro-Ecosystems

Annual Progress Report
October 1979-September 1980

Prepared by M. H. Smith, J. J. Alberts, D. C. Adriano
K. W. McLeod, J. E. Pinder III

Savannah River Ecology Laboratory

Prepared for
U.S. Nuclear Regulatory
Commission

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ABSTRACT

Our studies investigating the environmental behavior of radionuclides released into agricultural systems by a nuclear fuel chemical separations facility have advanced considerable within the past year. Manuscripts reporting uptake of Pu and Cm by agricultural plants have shown that Cm was taken up in greater quantities than Pu. Lime addition to soil depresses uptake of Cm while chelate addition to limed soil increases uptake of Cm. No conclusive effects of soil amendments were observed with regard to Pu uptake. Cesium uptake by agricultural plants differs between species, plant part and year of harvest, with an indication that cesium may be increasingly taken up with the passage of time in some instances. Soil amendments affect ^{137}Cs uptake similarly as that reported for ^{244}Cm.

Rice varities do not differ in their Pu or Cm uptake, nor does uptake by rice in flooded soils differ from those uptake rates reported for dry-land agricultural species. Curium and $^{239,240}Pu$ appear to be equally available to rice and more available than ^{238}Pu.

Broadleaf crops appear to exhibit Pu concentrations dependent on their shape and form, indicating the importance of physical processes in this field experiment.

Because of crop failures caused by drought conditions, there has been little progress on our U-field studies.

This report is prepared to indicate research highlights in the Savannah River Ecology Laboratory's (SREL) research program for the Nuclear Regulatory Commission. It is provided for information only and is not to be considered as formally published literature. We request that no citation be made of information contained herein without express consent of the investigator.

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SREL is a regional ecology laboratory operated by the University of Georgia for the United States Department of Energy (DOE). Our objectives are to carry out basic and applied research and to disseminate the findings through publications in the open scientific literature, technical reports, or other means of communication to the public, government, and scientific communities. General research programs at SREL include mineral cycling and aquatic stress. More specific programs include the fate of coal combustion contaminants and radioecology of actinide elements. The resident research staff consists of approximately 15 senior scientists and nearly 50 associated technical, clerical and support staff. Most senior scientists work in several research programs so that their expertise can be fully utilized by the entire staff. Emphasis is placed on field-oriented research that has broad ecological significance but that also relates to regional as well as local problems on the Savannah River Plant (SRP).

The Savannah River Plant (SRP) has the unique situation of having two nuclear fuel chemical separations facilities which have been in operation for twenty-five years. From these facilities, we have both low-level aerial and liquid waste effluents. These effluents differ in radionuclide composition due to the type of release (liquid or aerial) and the facility from which they emanate. To date, research funded by DOE, has concentrated on plutonium contamination of agricultural crops due to aerial effluents from H-Area. We have established a one hectare field and farmed this field for 4½ years. The other reprocessing facility on the SRP, F-Area, handles different types of fuels and consequently the composition of its effluent is different. For example, aerial releases of U-235,238 from F-Area in 1976 were 4.87 mCi, an order of magnitude higher than H-Area. This has been a typical yearly uranium release for the last twenty-five years (1). These releases have created a unique soil contaminated by low level aerial effluents from operating separations facilities over a twenty-five year period. The present chemical form of the contaminants is due to natural weathering processes. Therefore, these contaminated soils could not be duplicated by artificial spiking or other weathering processes and represent a real world situation.

Our NRC research program has concentrated on experiments to determine the long term availability of radionuclides to agricultural plants from soils contaminated by a nuclear fuel chemical separations facility. In these experiments, we are also investigating factors which might modify availability of nuclear contaminants such as different crop species, soil amendments and soil types. A second phase of research examines the plutonium contamination of edible broadleaf crops grown in the vicinity of H-Area. The third phase of our research program is the establishment of an agricultural field at F-Area to conduct studies on the environmental behavior of U in agricultural systems. From these studies, we can determine whether U acts any differently in the environment than other actinides.

Progress in each of these areas of research will be discussed in the following sections. In addition, a number of other localities on the SRP in which U, Pu, Cm and other long-lived radionuclides have been deposited are being investigated to determine their appropriateness for

research regarding the environmental behavior of these nuclides. We feel that an expansion of the research program into field studies of other long-lived radionuclides emanating from operating nuclear fuel chemical separations facilities and of common concern in waste management is an appropriate direction. These types of studies would be also unique in that we would be able to study aged contaminants rather than spiked soils, which is much more indicative of real world situations.

RCOT UPTAKE OF ACTINIDE ELEMENTS BY AGRICULTURAL PLANTS

Much research on root uptake of actinide elements has been conducted at many different sites on soils which have been contaminated by weapons testing, (2, 3, 4), spiking (5, 6), or liquid wastes (7, 8), but information is available only from the SRP regarding the behavior of low-level aerial releases of these nuclides from separations facilities (9, 10, 11, 12, 13, 14).

Since its inception in the early 1950's, plutonium monitoring at source points on the SRP indicated a total of 3.6 curies released to the atmosphere, 13.9 curies to the seepage basins, and 0.4 curies to the streams. Most of the aerial releases occurred prior to 1970, but minor releases are still occurring. The nature of the spent fuels being processed assures that low level releases of Americium-241 and Curium-244 have occurred. Although these levels are lower than that for plutonium isotopes, they are still detectable with existing analytical techniques. Thus, the industrial operations at SRP provides an excellent research opportunity to study some of the critical pathways of the actinide elements (U-235, U-238, Pu-239,240, Am-241, and Cm-244) from typical agricultural operations in the southeastern United States.

Pathways of these nuclides to man from agriculture include inhalation and ingestion. Inhalation of soil-borne particulates arising from various phases of agricultural operations has been examined on the SRP previously (15). The hazard of ingestion, and on rare occasions inhalation, as in the case of tobacco smoking, of surface-deposited and/or internally deposited nuclides and inhalation of dust particles from processing contaminated food products, depend on the concentrations of transuranics in food products and the distribution of the total contamination between external and internal deposits. Internally deposited transuranics would most likely be ingested. Externally deposited transuranics would probably be resuspended by food reprocessing operations.

DOE funded research has provided data on the movement of Pu through these various pathways from crops grown in the vicinity of separations facilities at SRP. Conclusions from these studies are based on the assumption that a similarity of isotopic ratio between biotic and abiotic components indicates transfer between these components. Data from winter wheat and soybean crops suggest that current fallout from separations facility stack constitutes at least 90% of the total Pu contamination of these crops. Consequently, this resulted in plant/soil concentration ratios of approximately 10^{-3} for wheat grain, and soybean bean several orders of magnitude higher than commonly reported values for root uptake (10, 11). Apparently plant assimilation via root uptake is apparently only a minor source of contamination under field conditions (10, 11, 13, 14).

Although our data indicate that root uptake of Pu is small compared to surface contamination, Am and Cm have been shown to be more available for uptake than Pu (16, 17, 18). If soil amendments modify uptake under field conditions, they could cause elevated concentrations in food

products. Furthermore, transuranics that have been incorporated into plant tissues may be chemically bound to proteins, lipids and other organic compounds. Gut transfer of these assimilated compounds may be greater than the current ICRP values determined using solutions of inorganic compounds. Thus it is very important to examine the internally fixed contaminants. This research attempts to answer the following questions:

1) Do agricultural species differ in their ability to take up radionuclides?

2) Do common soil amendments change the availability of the radionuclides?

3) Do radionuclides increase in availability with time of residence in the soil?

To accomplish these objectives a long-term study of plant uptake under greenhouse conditions using soils contaminated by releases from nuclear fuel chemical separations facilities has been undertaken. The shelter of a greenhouse is necessary because surface contamination of vegetation by global fallout and SRP aerial releases can obscure root uptake under field conditions.

In the past year, we completed the fourth year of this experiment. The radionuclide analysis of the samples from the first year's harvest are complete. These results are to be reported in two manuscripts. The first manuscript, reporting on the uptake of curium by these agricultural crops, has been accepted for publication in Health Physics. The second manuscript, reporting on the plutonium uptake, will appear in an issue of Soil Science, in which A. Wallace of UCLA has arranged for publication of NRC supported research. Copies of these manuscripts are included in their entirety in Appendixes A and B.

The results from the first year of this study partially answer the first two objectives of this study. In brief, due to difficulties with many samples being at or below detection limits, it is difficult to make any strong conclusions. It appears that neither plant species nor soil amendment strongly modify uptake rates. Some differences do occur in Cm uptake (lime depresses uptake, chelate addition to limed soil increases uptake, clover uptake rate greater), but none of these observations can be tested statistically for Pu uptake due to small number of samples and detection limit difficulties. Radionuclide analyses of the harvests from the fourth year of this experiment have begun. These results will be used to examine the third objective, i.e. increase in availability with time, and to re-examine the first two objectives. A difficulty in experimental design in that we did not have adequate greenhouse space to examine chelate addition separately will be rectified as soon as we are able to move into our new greenhouse.

ROOT UPTAKE OF CESIUM-137 BY AGRICULTURAL CROPS

Cesium-137 is a very common radionuclide released as a fission product from the nuclear industry. On the SRP, during the period from 1954 to 1976, 602 Ci of ^{137}Cs were released into streams (1). The floodplain soil described in the previous section on actinide uptake was found to have approximately 300 pCi ^{137}Cs/g (dry weight). No other gamma emitters were found in any significant quantities in this soil. Therefore gamma analysis for ^{137}Cs was conducted on the samples from the "actinide uptake" experiment prior to alpha counting.

The importance of cesium contamination of human foodstuffs has long been recognized due to its ability to substitute for potassium, a required element in the human diet. Thus factors that affect cesium uptake by agricultural plants could easily affect that reaching man through the foodchain. For the sake of brevity, the methods will not be described here but can be found in Appendix A. Only the floodplain soil had enough ^{137}Cs to justify counting the samples. Counting was accomplished utilizing a Nuclear-Chicago 400 channel pulse height analyzer with a 5.08 cm NaI well type detector.

Results

Due to the relative simplicity in counting gamma emitters as opposed to alpha emitters, data from several years of harvests of this uptake experiment are available. Thus trends caused by temporal as well as treatment and species differences can be examined.

Corn. - Of the vegetative portions of the corn plant, leaves had consistently higher concentrations of ^{137}Cs than stems (Figs. 1 and 2). The effect of liming of the soil was to decrease ^{137}Cs uptake. Chelate addition to limed soil produced little difference in uptake. Cesium concentration in leaves and stems were greater in the first year than the second or third year, which were generally similar.

Tassels from the corn plants had high Cs concentrations, similar to those found in corn leaves (Fig. 3). The effects of soil amendments and time were similar to that observed in the vegetative portions. Only in 1978, the second year of the experiment, were enough kernels produced to warrant analyses. Cesium concentrations in this tissue were low (Fig. 4). Limed decreased Cs concentrations in this tissue, while chelate addition appeared to totally counteract the depressive effect of lime.

Wheat. - At the time when wheat grain is ready for harvest, there exists very little erect vegetative material except for stems. Therefore cesium concentrations were not determined for vegetative material but only determined for wheat grain. The basic trends in soil amendment and time effects which were observed for corn are also apparent in wheat grain (Fig. 5). The range of Cs concentrations for wheat grain is also very similar to that for corn kernels.

Soybean. - Soybean is similar to wheat in regard to little vegetative tissues remaining on the plant when the fruit has matured. In order to obtain Cs concentrations for leaves, we sampled a limited number of

leaves prior to defoliation of the plants in the 1979 growing season. These leaves and associated stems showed very similar Cs concentrations (Figs. 6, and 7). Effects of soil amendments were very similar to those described previously for corn and wheat. The effect of time is observed to act in the opposite manner as that in corn. The Cs concentrations of stems is tending to increase as the number of times soybean is grown on the same soil.

Reproductive tissues (hulls and beans) show opposite effects of time on Cs concentrations, with the pattern in beans being similar to that shown by corn (Fig. 8) while the hulls being similar to that of soybean stems (Fig. 9). The effects of soil amendments continue to show a decreased uptake following lime addition and very little effect of chelate addition to limed soil.

Bahia Grass. - The effect of soil amendments on the Cs concentration of bahia grass is very strongly exhibited (Fig. 10). Lime depresses the Cs concentration by a factor of 7. Chelate addition to limed soil continues to only exhibit a very slight effect. Concentrations in plants grown in non-amended soil remain high (~ 2100 pCi/g) for two years before they begin to decline. Time appears not to be a factor in uptake from amended soils. These data represent analysis of composites of two or three harvest periods per year. Since multiple harvests per year are possible with bahia grass, we also have the ability to examine time effects within growing seasons (Fig. 11). If one combines all three years and examines concentration vs. month, the Cs concentration increases from May to August and then begins to decline. This is very similar to the growth pattern of bahia grass and may simply reflect this growth activity. All three soil treatments respond in this same manner. Thus the pattern of Cs concentration within a single year may be simply due to when the sample was taken which was dependent on the growth pattern of the species. There was also low Cs concentrations of the initial harvest, probably due to incomplete root exploitation of the soil volume within the pot.

Clover. - Soil amendments appear to have affected Cs uptake for only the first year's harvest (Fig. 12). Amendment effects within the first year are identical to those already described. Following the first year, the amendments appear to have little effect with plants in all soil treatments having similar Cs concentrations. These concentrations are lower than those observed for plants from the first year's harvest when grown in non-amended soil. Information similar to bahia grass can also be determined from the clover crop due to multiple harvests per year. Clover has a different yearly growth pattern than bahia grass. Clover is much more a cool season crop. No particular response can be observed in Cs concentration of clover (Fig. 13).

Discussion

A consistent pattern in the effects of soil amendments can be observed from these data. Lime addition, which increases the soil pH, has resulted in decreased ^{137}Cs uptake. As lime addition has frequently been shown to depress metal uptake, this result was anticipated. Since

chelate addition has been shown to increase metal uptake, the fact that the chelate addition did not more fully counteract the lime effect is somewhat surprising.

Of the vegetative tissues, bahia grass had considerably greater cesium concentration ($\sim$ 2100 pCi/g) than any other species, followed by corn leaves ($\sim$ 700 pCi/g) and clover ($\sim$ 375 pCi/g). Corn and soybean stem had less than 225 pCi/g. Of the reproductive tissues, corn tassels had $\sim$ 420 pCi/g while soybean hulls had $\sim$ 225 pCi/g. Of course those tissues are not consumed. Soybean beans had more cesium than either corn kernels or wheat grain. These tissues are actually consumed and represent a potential dose-to-man via the ingestion pathway. These species do appear to have different rates of cesium uptake. If one considers only the vegetative tissues, the grasses (bahia grass and corn) have higher rates of uptake than the other crops. This does not hold true for the reproductive (edible) tissues.

The temporal effect also seems to differ between species. Several patterns are apparent. First, a continual decrease in Cs uptake rate for all three years (e.g. corn leaves and tassels, wheat grain, soybean beans, and clover). This pattern might also be interpreted as having no change between the second and third years, but definite decrease from the first year to the second. A second pattern is found in bahia grass which shows similar uptake rates for the first two years and then a decrease in the third year. Thus the pattern of an overall decline in the three years is similar to the first pattern. The third pattern is observed only in soybean stems and hulls, but illustrates an opposite pattern. Cesium concentrations increase in each of the three years. This pattern is of most concern as it represents a potential problem of increasing availability of a potential contaminant. Why these three different patterns exists in these crops is unknown. The patterns are not consistent within species or within a type of tissue. For now they must remain unexplained.

Due to the limited Cs uptake from amended soils, it is difficult to observed whether the temporal patterns also exist after the soil has been amended.

Analysis of these tissues grown during the 1980 growing season will assist in our determination of the consistency of the observed patterns in species, soil amendment and temporal effects. These results will be available during the next year.

UPTAKE OF ACTINIDES BY RICE

Rice is the most important food crop to billions of people in the world. It is grown under a wide variety of climatic and environmental conditions (temperature, rainfall, soil aeration). For most varieties, at least a portion of the growing season finds the rice crop inundated by water. In fact in the United States, rice production only occurs in bottomlands in close proximity to large rivers. The floodplain areas, whose soil is being used to conduct our other uptake studies, would be a suitable environment and soil for rice production. Therefore we sought to determine the Pu and Cm uptake rates for four widely used varieties of rice.

Materials and Methods

Four varieties of rice (Oryza sativa L.) were used in this study. Three varieties (Belle Patna, Nato, and Starbonnet) are commonly used in the United States. A fourth variety, IR-1561 developed by the International Rice Research Institute (IRRI), is widely used in Asia. Floodplain soils as described in Appendixes A and B were used. In this soil are significant concentrations of Pu and Cm, which were deposited on the floodplain as a consequence of the operation of a nuclear fuel chemical separations facility. These radionuclides have been in the environment for long periods of time and have derived their present state through natural weathering processes. It was necessary to move the soil to the greenhouse for the uptake experiment for several reasons. First and foremost is the fact that the floodplain is currently forested and light would be limiting for good field crop production. Also shelter from current releases would be required to restrict plant concentrations to only those due to root uptake and translocation. Pots contained 5 kg of soil with six replicates of each variety used. The pots were continually flooded from germination until harvest. At harvest, the plants were clipped above the high water mark and separated into foliage and grain. Dry weights were obtained, followed by ashing for actinide analysis. Determination of ^{238}Pu, $^{239,240}Pu$, and ^{244}Cm concentrations were conducted by LFE Environmental Analysis Laboratories by total dissolution and low level alpha spectrometry methods.

Results

Soil concentrations of these actinides differed slightly from previous analysis on a similar batch of soil. Plutonium soil concentrations were 7.432 and 0.056 pCi/g for ^{238}Pu and $^{239,240}Pu$, respectively. Curium-244 soil concentration was 0.387 pCi/g.

Concentration of ^{238}Pu in rice foliage were approximately 2 fCi/g (Table 1) with little variation between varieties (1.91 to 3.67 fCi/g). Lower concentrations of $^{239,240}Pu$ were found in the foliage, but there was also less in the soil. Concentrations of this radionuclide ranged from 0.42 to 1.40 fCi/g. Curium-244 concentrations were similar for the foliage of all four varieties (0.68 to 0.89 fCi/g).

The concentrations of grain (Table 1) of all three radionuclides were generally lower than for the foliage (Range : 0.67 to 2.66 fCi/g for ^{238}Pu; 0.44 to 0.86 fCi/g for $^{239,240}Pu$; and 0.51 to 1.73 fCi/g for ^{244}Cm).

Concentration ratios for these actinides were low (Table 2), with plutonium-239,240 concentration ratios of 10^{-2} to 10^{-3}, while ^{238}Pu were 10^{-4}. Since the plant concentrations were low and near the detection limit, the differences in soil plutonium concentrations could account for the slight differences in concentrations ratios between the two plutonium isotopes. Concentration ratios were also lower for grain than foliage.

Discussion

Plutonium and curium concentrations of rice foliage and grain grown on the floodplain soil were quite low (less than 4 fCi/g). Resulting concentration ratios were also quite low (10^{-5} to 10^{-2}) and very comparable to those concentration ratios observed under dry-land agriculture. There does appear to be differences in uptake of the three radionuclides, with $^{239,240}Pu$ having the highest concentration ratio, followed next by ^{244}Cm and then ^{238}Pu. This result can also be shown by examining the ratio of discrimination ratios for the three radionuclides (Table 3). Using this method, $^{239,240}Pu$ is taken up $\sim$ 6 times greater than ^{244}Cm and $\sim$ 47 times greater than ^{238}Pu. Curium-244 is taken up $\sim$ 7 times greater than ^{238}Pu.

There appears to be no consistent differences between rice varieties in their ability to extract either Pu or Cm from the soil. Resultant dose-to-man via the ingestion pathway would appear to be minimal based on these results even for an average adult Asian diet where consumption would be approximately 165 kg/year.

PLUTONIUM CONTENTS OF BROADLEAF CROPS GROWN NEAR A NUCLEAR FUEL CHEMICAL SEPARATION FACILITY

Broadleaf crops represent a potentially very important pathway of plutonium to man, from our system, where aerial releases and resuspension represent the prime contamination modes. Since these crops have large amounts of tissue exposed to the atmosphere, there would be considerable quantities of plutonium continually being deposited on these tissues, which ultimately would be consumed. This is a very different case from the major field crops (wheat, corn, soybean) whose edible portion is buried deep within non-edible tissues. Thus without consuming nearly as much of the foodstuff as for a major food crop, a similar dose could be received due to greater Pu concentration in the broadleaf crops.

Since the plutonium released from the stack is associated with dust particles and these particles are transported largely by physical processes, they can be dislodged from leaf surfaces and eliminated from the foodstuff. Therefore, in addition to examining the Pu concentration of these broadleaf crops, it is also important to examine common food cleansing techniques to determine their ability to remove Pu from the edible portion.

We have concentrated our efforts in determining the Pu concentration contributed by a nuclear fuel chemical separations facility to four common broadleaf crops grown and consumed in the southeastern United States. The efficiency of food preparation techniques in removing surface deposited Pu was also examined.

Materials and Methods

These studies were conducted in the H-Area vicinity, where chronic low-level Pu releases are occurring. A field, which has previously been used for other agricultural studies, was located 420 m northwest of the stack, the point of Pu release. A 4 x 4 Latin Square experimental design was used with 3 rows of the crops within each block. Each plot was 6 m x 4 m. The field layout is shown in Fig. 14. Four broadleaf crops ((1) turnip greens, Brassica Rapa L.; 2) cabbage, B. oleracea var. capitata L.; 3) broccoli, B. oleracea var. botrytis L.; and 4) lettuce Lactuca sativa L.) were grown in the spring of 1978. Broccoli, lettuce, and cabbage were transplanted into the field site as small plants, while turnips were seeded directly. Upon maturation of the crops, they were harvested and portions cleansed in manner similar to what would be done prior to consumption. Control samples, which were grown off the SRP, were obtained for all crops from a local farmers' market except broccoli, which was unavailable. All samples were dried, weighed and ashed prior to Pu analysis. Current deposition rates were determined by tacky paper collectors placed 7.6 m above the field. Each paper collector had surface dimensions of 33.0 x 25.4 cm and was replaced weekly. A total dissolution and alpha spectrometry were used to determine Pu concentrations.

Results and Discussion

The results reported here are preliminary and represent only a portion of the analysis. Determination of Pu concentrations is continuing and will be completed soon.

Since a semi-head variety of lettuce was used in this study, there was the possibility of aerial released plutonium filtering down into the head. Therefore some heads were washed as a head and some washed after the leaves had been separated, in addition to whole unwashed heads. Unwashed heads contained the highest concentrations of both ^{238}Pu and $^{239,240}Pu$ for lettuce samples (Table 4). Plutonium concentrations were decreased by half or more by washing the lettuce. All lettuce samples from the field near H-Area contained more Pu than the off-site control sample, indicating a contribution by the separations facility.

Turnips also showed higher Pu concentrations when grown near H-Area than off-site, but the Pu concentrations were not as high as those found in lettuce. Very few of the cabbage samples have been completed, but those that have show very minimal Pu concentrations in washed cabbage heads. These concentrations are much lower than comparably treated lettuce samples indicating the possible importance of the solid head form of cabbage as opposed to the semi-solid head of lettuce.

Broccoli represents an edible aerial crop but differs from the other three broadleaf crops in that the edible portions of broccoli is actually stem and floral tissue, whereas lettuce, cabbage and turnip greens are leaf tissue. Thus edible broccoli tissue is exposed to the aerial effluent only for a short time as opposed to the other broadleaf crops. Washed broccoli showed higher concentrations then unwashed samples. The reason for this is unknown at present but may be clarified with the completion of the analysis.

Of the three leafy broadleaf crops, comparable samples of each showed highest Pu concentrations in lettuce followed by turnip and cabbage. This may purely be a function of the manner in which that species grows to maturity. For example, lettuce (our semi-head variety) was continually open to aerial deposition and once the Pu was deposited the possibility of the Pu being washed out of the head was very low. Turnip greens represent an intermediate case of large individual leaves being exposed to both aerial deposition and the high probability of the material being washed off by rainfall. Cabbage then represents a case of a crop growing inside of a protective cover. The oldest leaves are on the outside of the head with new growth occurring inside these old leaves. Thus most of the head is never exposed to the aerial releases of plutonium and consequently low Pu concentrations were observed.

Since the concentration of plutonium in these crops was much greater than that expected by the assumed concentration ratios for root uptake exclusively, this transfer mechanism was not a major pathway of plutonium to these crops. Therefore, due to the situation that the soil contains more $^{239,240}Pu$ than ^{238}Pu and the opposite being true for the current aerial effluent, we can identify the importance of the contami-

nation pathways of examining the ratio of Pu isotopes in the plant samples. Since this ratio is near 1.0, both contamination pathways (direct deposition of current releases and resuspension of surface soil) were occurring and apparently in approximately equal proportions. With the completion of the Pu analysis we will be better able to determine the relative importance of these two pathways.

Uranium Field-Crop Studies

Studies of the comparative environmental behavior of uranium released from a nuclear fuel chemical separations facility have been in progress for two years. The behavior of U will become much more important to understand if we shift to a U-Th fuel cycle. Aerial releases of total U from the F-Area facility are 1.50 $pCi/m^2/wk$ with over 99% of the total U being U-238. This has resulted in soil concentrations of total U of 0.6 pCi/g of soil at a 550 m distance from the stack. This is a very similar situation to our H-Area study site in which Pu has been studied and will offer an excellent opportunity for comparative studies.

In the first year, a winter wheat crop was planted and harvested in a field in close proximity to F-Area. The field was cleared of woody vegetation and common agricultural practices followed (plowing, discing, liming and fertilization). Growth was very satisfactory. On an adjacent area of this same field, we put in, by the same methods, a cover crop of winter rye to help stabilize the soil. This will allow a comparative study between the two species. In the second year, we attempted to raise corn and soybean crops in a similar fashion to that described for wheat and rye. Due to deer browsing and the exceptional drought which we experienced last summer in South Carolina, the crops were total failures. We will be raising these crops during the coming year, with precautions taken to avoid deer and drought problems.

In conjunction with this crop production, we sample the soil with a split barrel corer both before and after preparation of the field to allow examination of U migration. Also, we have erected tacky paper collectors from which can be estimated the amount of resuspension occurring from the surface soil. The addition of a second and third tacky paper, one on the underside of the collector platform and one suspended 1 m above the ground will allow better estimates of the resuspension component.

In order for us to determine the relative importances of the possible pathways of U to the crop, we have also grown these crops in pots in non-contaminated soil at the field site. These pots are isolated so that they only receive U from direct deposition and the resuspension and root uptake pathways are eliminated. Also, small quantities of the contaminated soil were taken to the greenhouse, where we are growing the same crops in pot culture so that we can determine how much of the uranium contamination of the field crops is due to root uptake. This will then allow us to determine how much contamination may be due to foliar deposition and retention as opposed to root uptake and translocation. Soil, vegetation, and tacky paper samples will be analyzed for U by leaching, or total dissolution of the samples and alpha spectrometric analysis.

We are currently working with LFE Environmental Analysis Laboratories on the analytical aspects of these studies.

SUMMARY

Our greenhouse studies designed to investigage the root uptake of transuranic elements by agricultural crops from soils contaminated by a separations facility and factors which affect their availability in the soil have made considerable progress within the last year. Complete results are available from the first year of this experiment and two manuscripts have been prepared reporting these data. Uptake of plutonium from soil is very low (concentration ratio is 10^{-3} or less). Neither lime nor chelate addition caused any large differences in uptake of Pu. Uptake of curium from soil is greater than plutonium and lime decreased uptake. Chelate addition to limed soils increased uptake above that observed from only limed soils. Conclusions based on this study indicate that only very minimal quantities of plutonium are incorporated into edible portions of crops. This would pose little health hazard due to consumption of contaminated foodstuffs. Curium may pose more of a problem due to greater availability from soil and possible greater gut-absorption coefficient.

These crops have continued to grow well and we have completed the fourth year of this experiment. Actinide analysis of these samples is currently in progress so that we might examine any temporal effect on root uptake of actinides by these crops.

This study has also shown soil amendment effects on cesium uptake similar to those observed for curium. The temporal pattern of cesium uptake is also very interesting in that different species and plant parts respond differently and that uptake rates of ^{137}Cs may increase over time.

Curium and plutonium uptake by rice grown under inundated soil conditions is very similar to that observed for our dry-land agricultural crop studies. Different varieties of rice show no obvious differences in uptake of these radionuclides. Grain and foliage do not differ in concentrations.

The preliminary results from the broadleaf crop experiment show that plutonium is transferred to those crops via both the direct deposition and resuspension pathways. Plutonium concentration varies based on plant morphology and growth pattern of the crops, but can be decreased from the ultimate foodstuff by common kitchen preparation methods.

The uranium field crop studies have been set back due to the failures of the soybean and corn crops caused by severe drought conditions. Precautions have been taken to avoid these problems in the future. Due to the large backlog of sample analysis from previous years, uranium analysis has not been started on either the wheat or rye crops, but will begin in the next year.

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Table 1. Actinide concentrations (fCi/g) of varieties of rice grown in flooded contaminated soils.

		^{238}Pu	$^{239,240}Pu$	^{244}Cm
Bell Patna	Foliage	2.28	0.42	0.85
	Grain	0.67	0.44	0.67
IR-1561	Foliage	1.95	1.40	0.89
	Grain	2.66	0.86	0.51
NATO	Foliage	3.67	0.98	0.81
	Grain	1.92	0.48	0.72
Starbonnet	Foliage	1.91	0.59	0.78
	Grain	0.76	0.58	1.73

Table 2. Concentration ratios of actinides in four varieties of rice grown in flooded contaminated soils.

		^{238}Pu	$^{239,240}Pu$	^{244}Cm
Bell Patna	Foliage	3.4×10^{-4}	9.0×10^{-3}	2.4×10^{-3}
	Grain	9.8×10^{-5}	9.4×10^{-3}	1.9×10^{-3}
IR-1561	Foliage	2.5×10^{-4}	2.0×10^{-2}	2.5×10^{-3}
	Grain	3.4×10^{-4}	1.2×10^{-2}	1.5×10^{-3}
NATO	Foliage	4.7×10^{-4}	1.8×10^{-2}	1.8×10^{-3}
	Grain	2.5×10^{-4}	8.9×10^{-3}	1.6×10^{-3}
Starbonnet	Foliage	2.6×10^{-4}	1.1×10^{-2}	2.0×10^{-3}
	Grain	1.1×10^{-4}	1.1×10^{-2}	4.6×10^{-3}

Table 3. Discrimination ratios[1] of actinides by rice grown in flooded contaminated soils.

		$^{238}Pu/^{239,240}Pu$	$^{239,240}Pu/^{244}Cm$	$^{238}Pu/^{244}Cm$
Bell Patna	Foliage	0.04	3.40	0.14
	Grain	0.01	4.65	0.05
IR-1561	Foliage	0.01	10.87	0.11
	Grain	0.02	11.54	0.28
NATO	Foliage	0.03	8.57	0.23
	Grain	0.03	4.53	0.14
Starbonnet	Foliage	0.02	5.50	0.13
	Grain	0.01	2.22	0.02

[1] Discrimination Ratio = $\frac{\text{concentration ratio for isotope a}}{\text{concentration ratio for isotope b}}$

Table 4. Plutonium concentrations (fCi/g) and isotopic ratio of plutonium radionuclides of broadleaf crops grown near H-Area.

		^{238}Pu	$^{239,240}Pu$	$^{238}Pu/^{239,240}Pu$
Lettuce	Control[1]	1.99	14.34	0.14
	Washed After Separated	82.68	88.52	1.008
	Washed As Solid Head	31.89	95.73	0.48
	Unwashed	174.93	246.01	0.80
Turnips	Control[1]	7.39	8.490	0.87
	Washed	77.31	61.02	1.33
Broccoli	Washed	9.84	39.32	1.73
	Unwashed	5.05	1.99	2.63
Cabbage	Control[1]	-	-	-
	Washed	1.52	0.52	3.86
	Unwashed	-	-	-

[1]Control samples obtained from local farmers' market

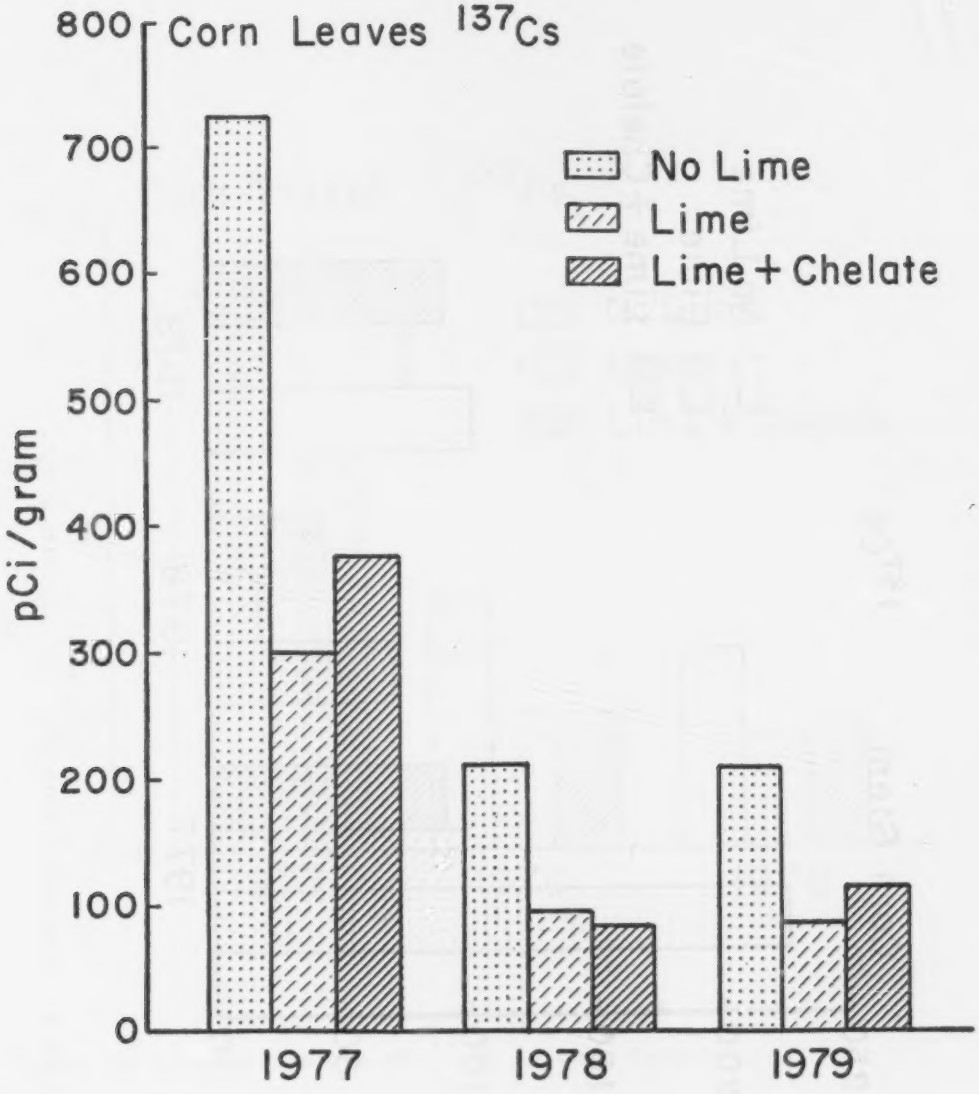


Figure 1. Concentrations of ^{137}Cs in corn leaves.

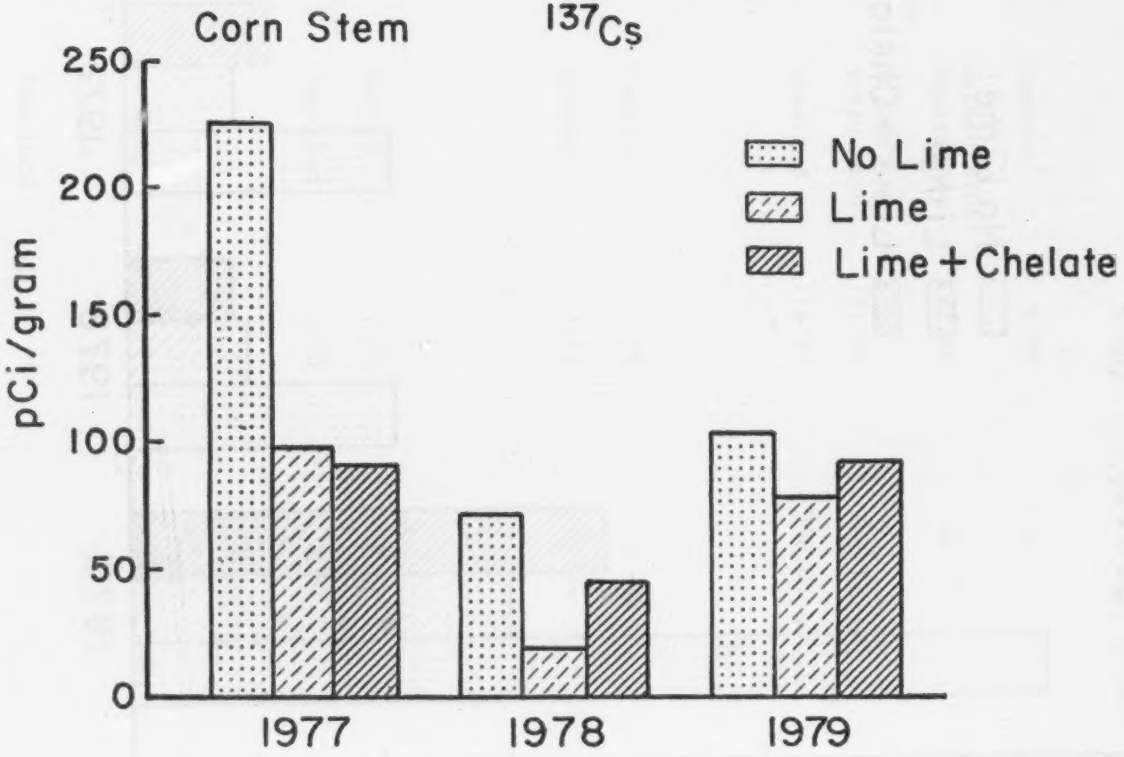


Figure 2. Concentrations of ^{137}Cs in corn stems.

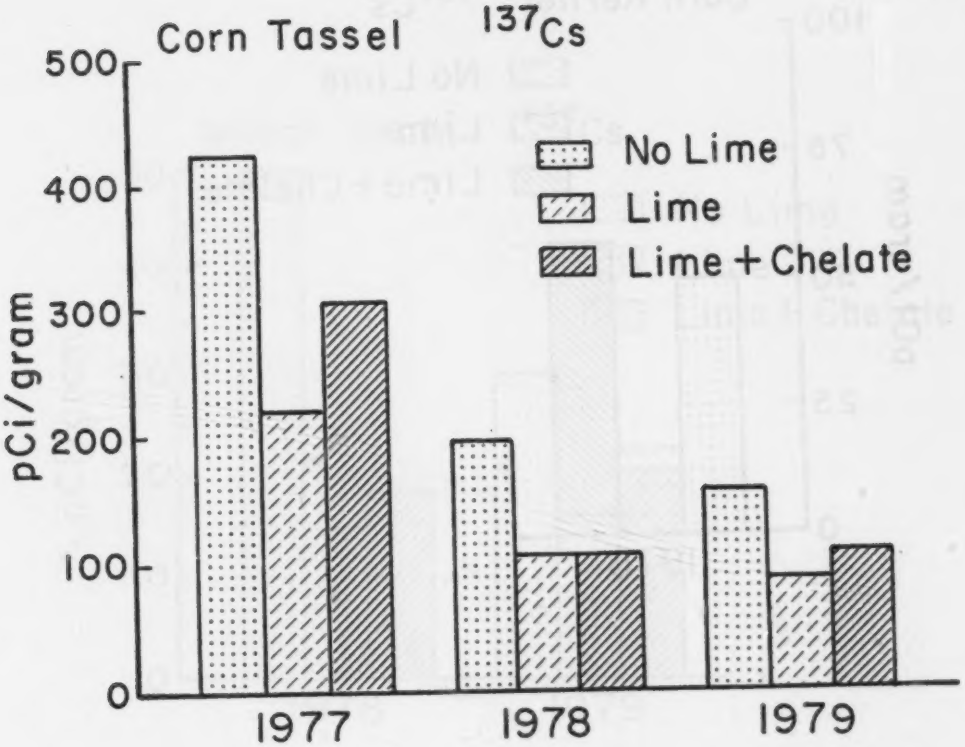


Figure 3. Concentrations of ^{137}Cs in corn tassels.

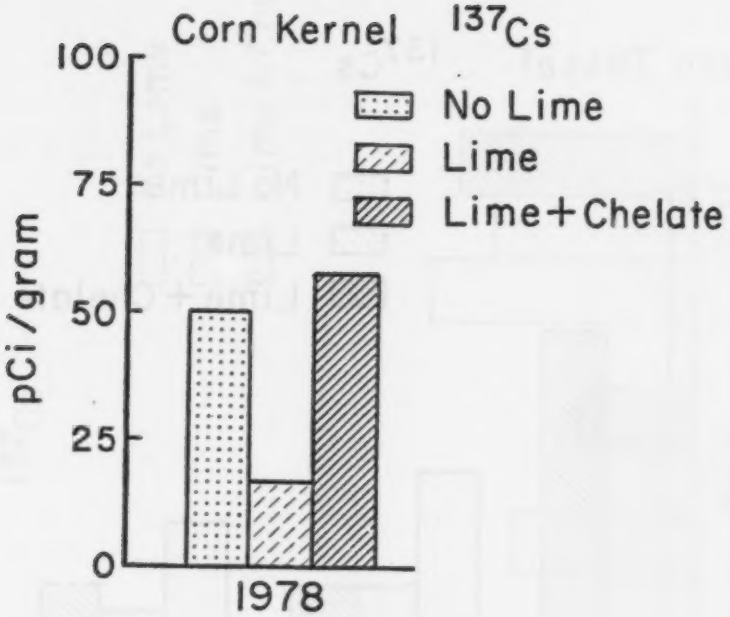


Figure 4. Concentrations of ^{137}Cs in corn kernels.

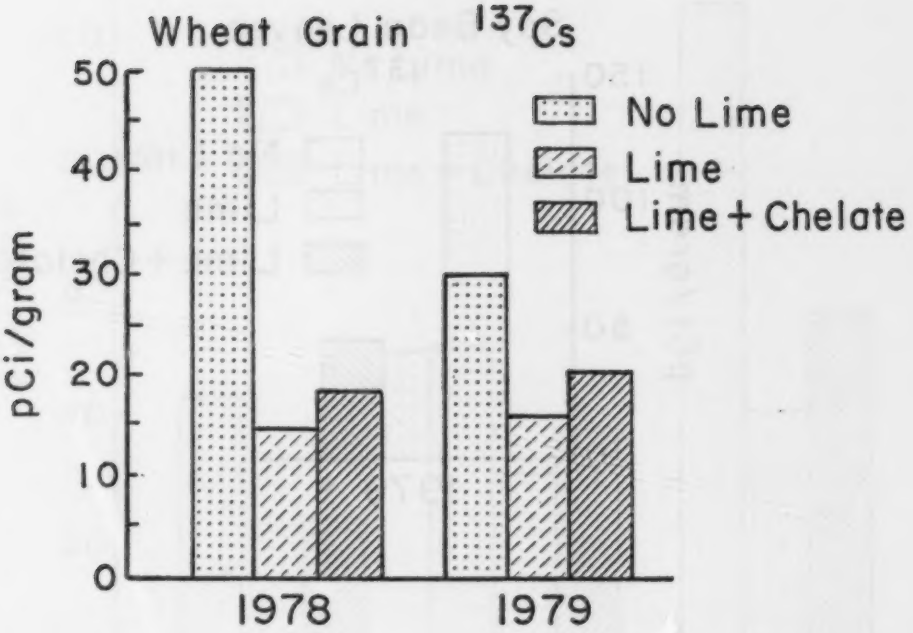


Figure 5. Concentrations of ^{137}Cs in wheat grain.

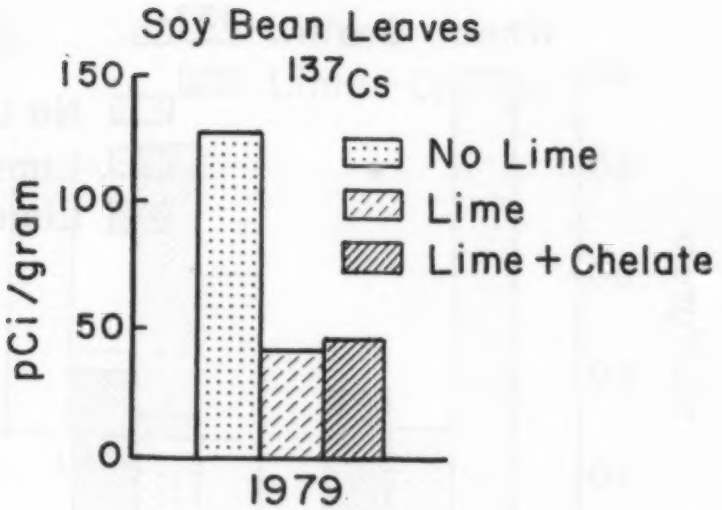


Figure 6. Concentrations of ^{137}Cs in soybean leaves.

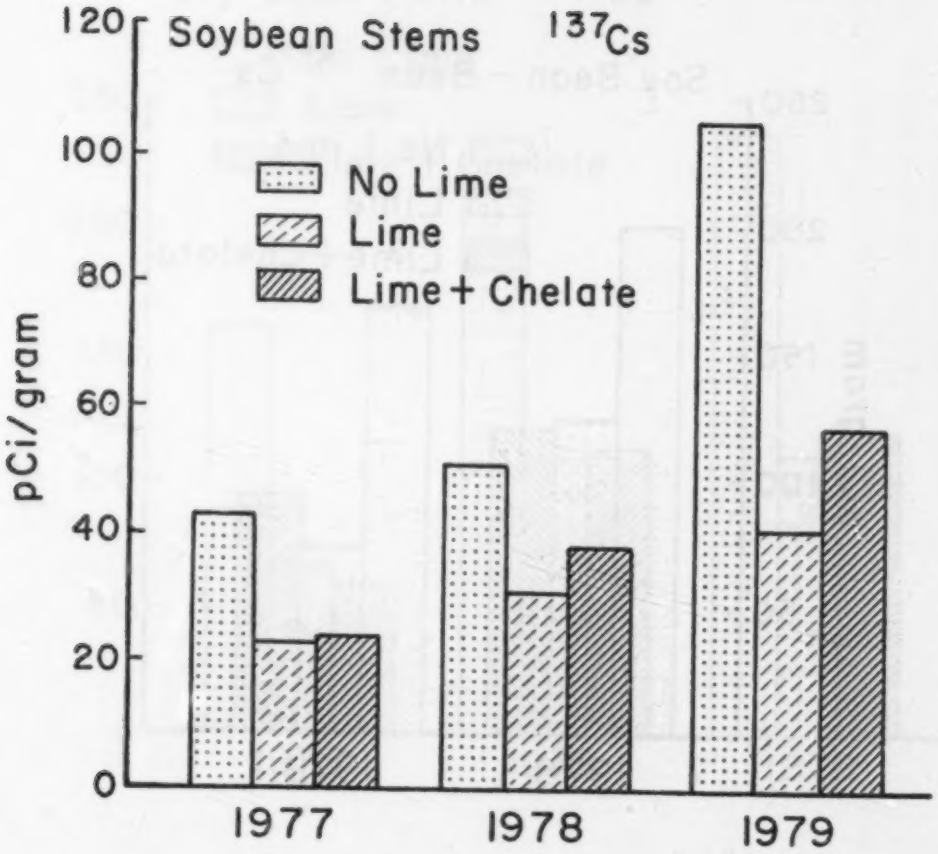


Figure 7. Concentrations of ^{137}Cs in soybean stems.

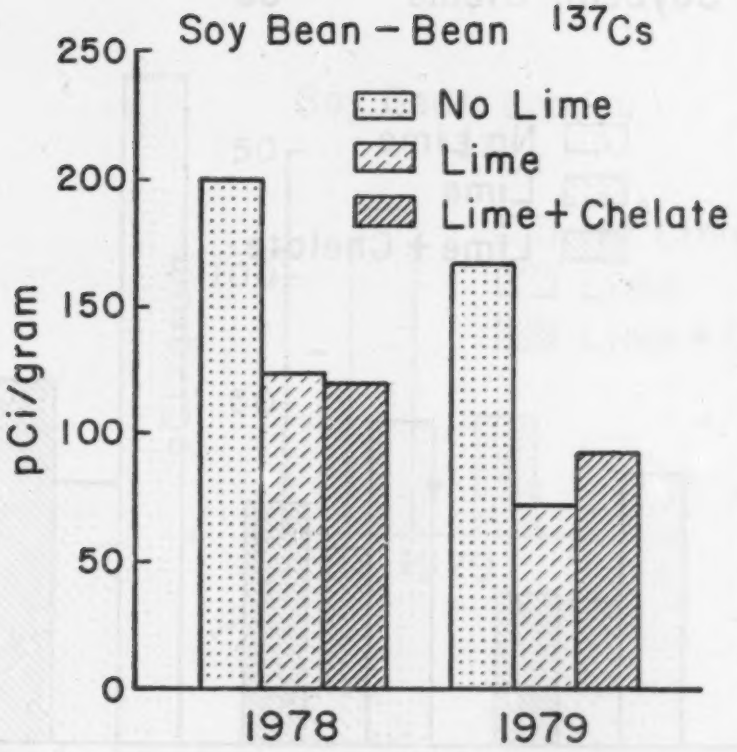


Figure 8. Concentrations of ^{137}Cs in soybean beans.

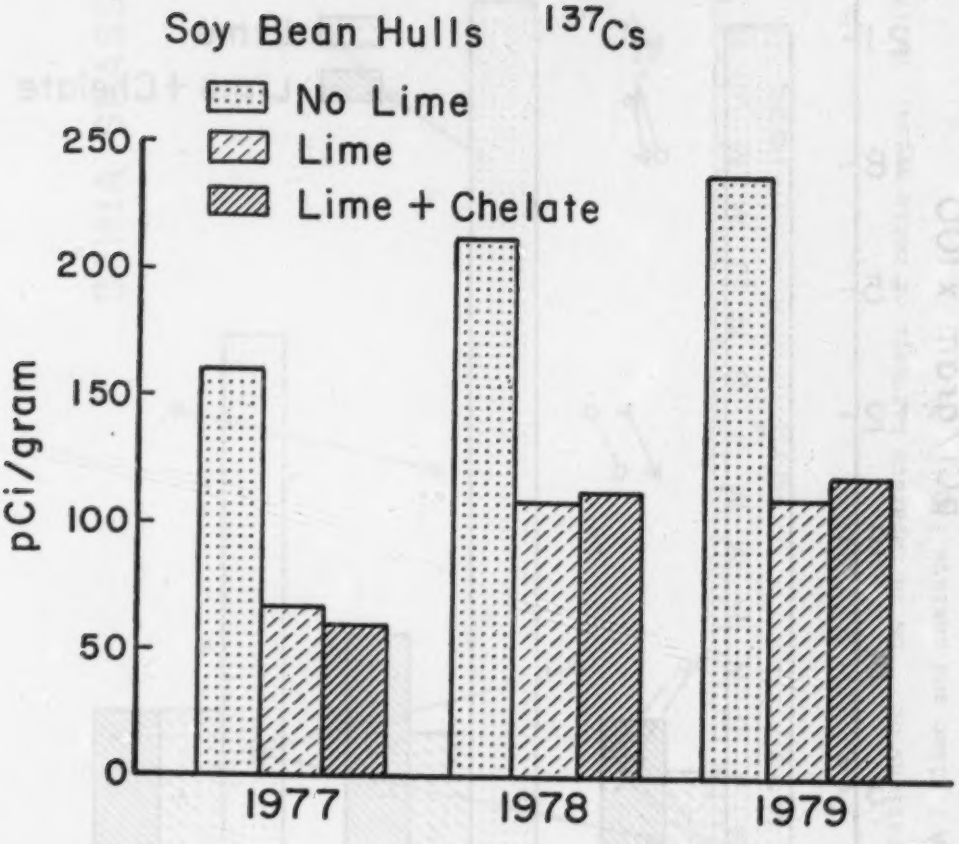


Figure 9. Concentrations of ^{137}Cs in soybean hulls.

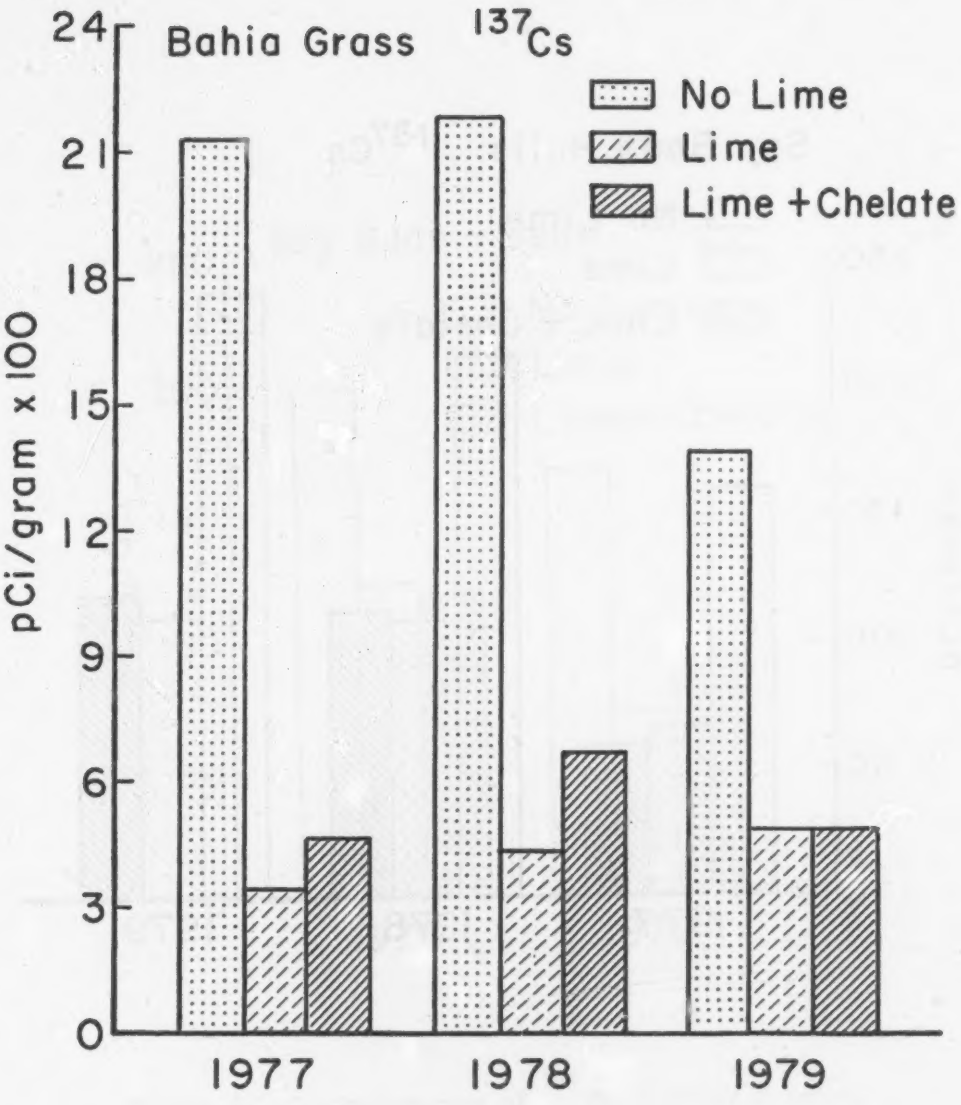


Figure 10. Concentrations of ^{137}Cs in bahia grass.

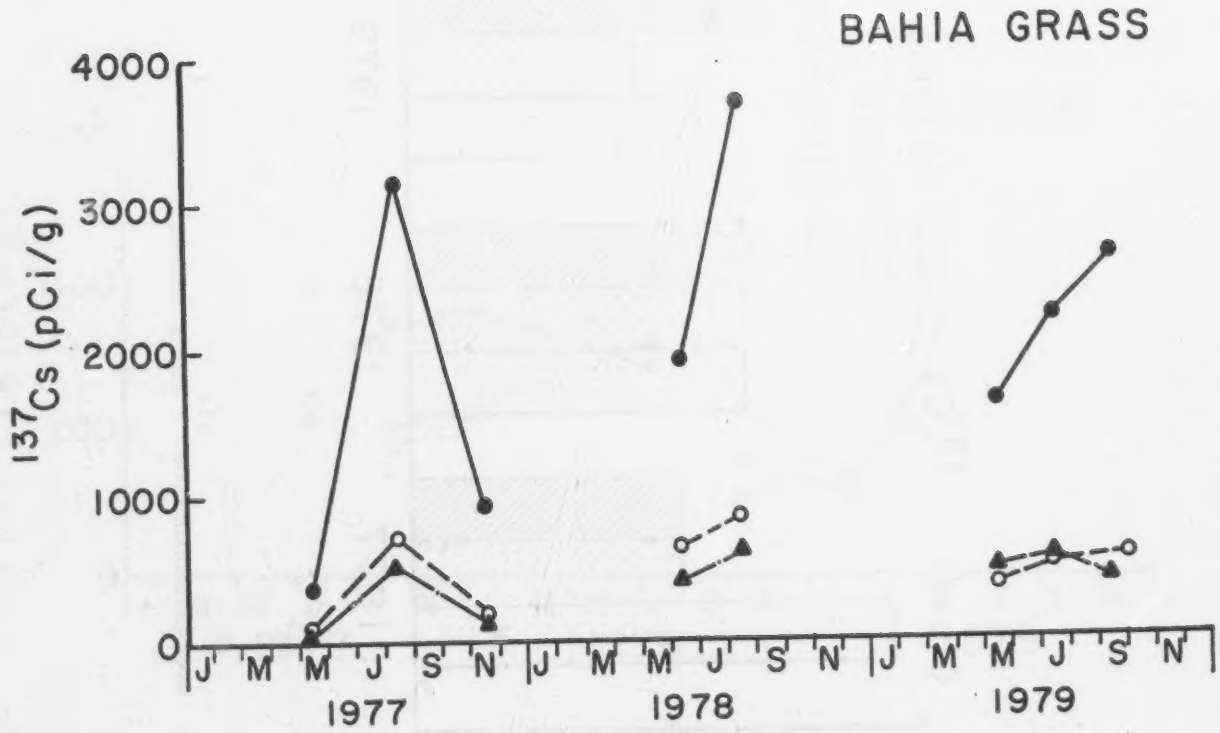


Figure 11. Concentrations of ^{137}Cs in separate harvests of bahia grass. No lime (●), lime (▲), lime and chelate (O).

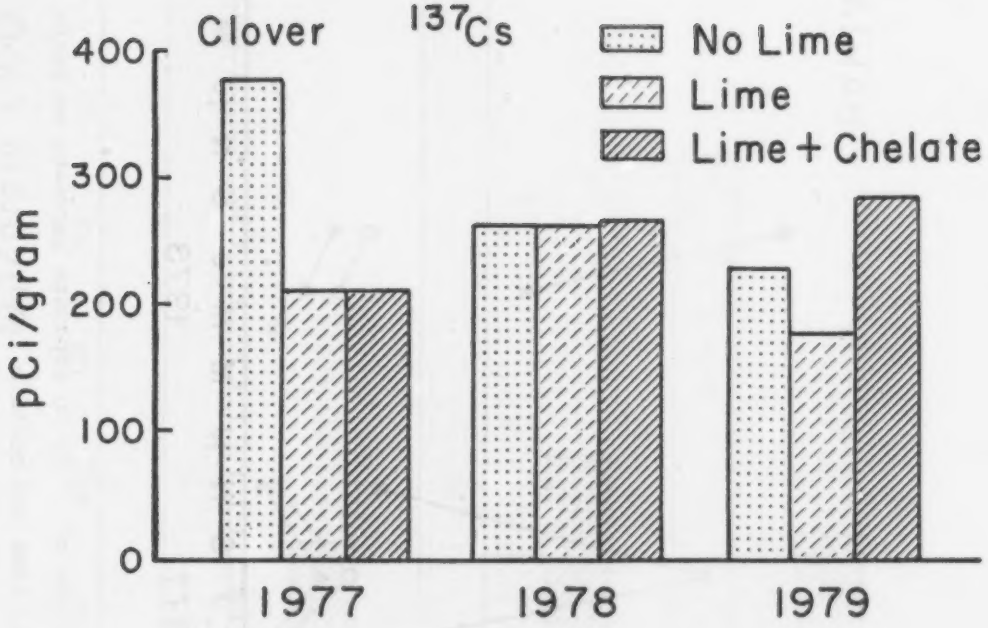


Figure 12. Concentrations of ^{137}Cs in clover.

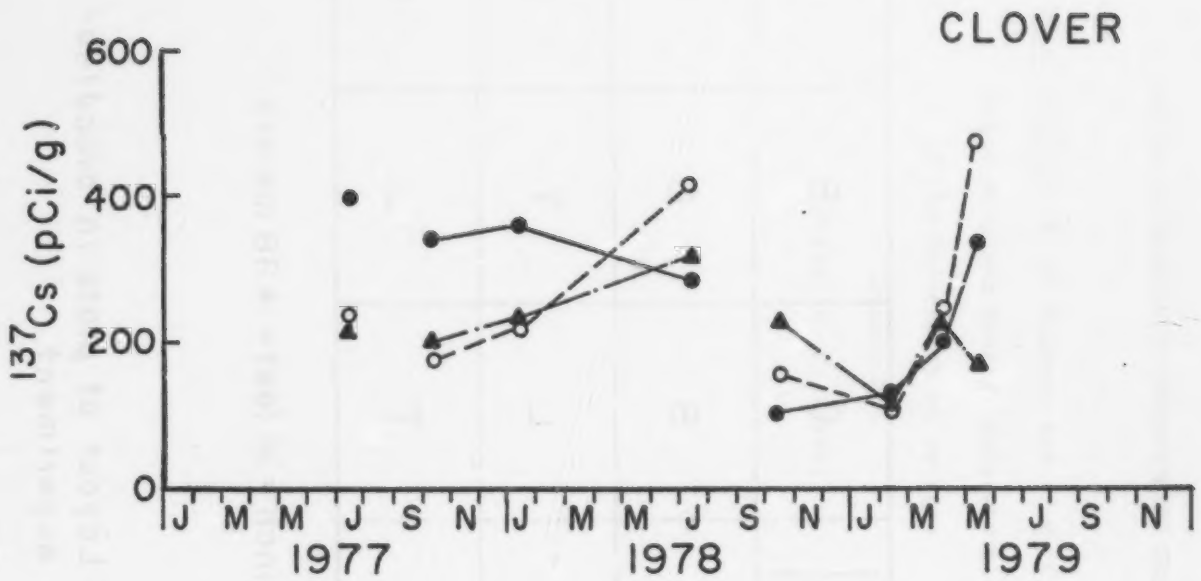
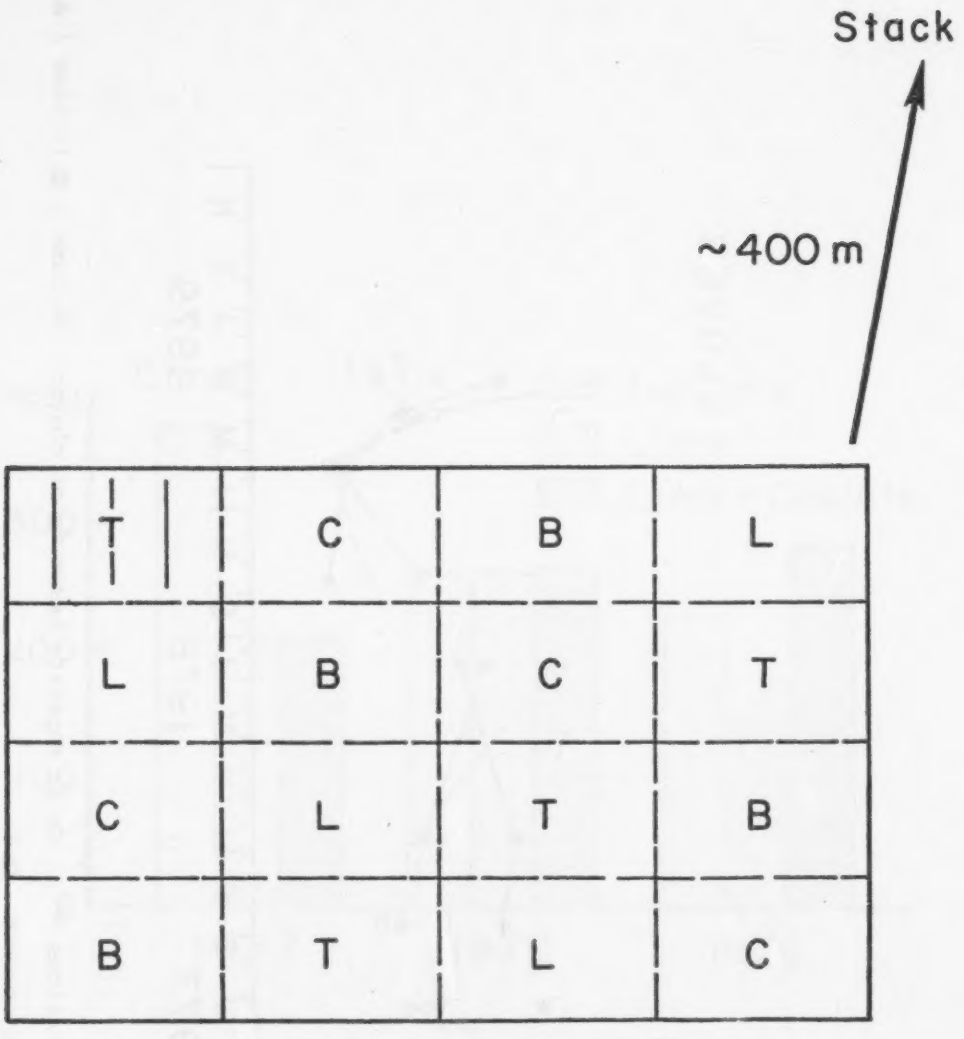


Figure 13. Concentrations of ^{137}Cs in separate harvests of clover. No lime (●), lime (▲), lime and chelate (O).



Scale: 1 inch = 16 feet = 4.88 meters.

Figure 14. Layout of plots in broadleaf crop field experiment.

CURIUM UPTAKE BY CROPS FROM NATURALLY-WEATHERED CONTAMINATED SOIL

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ABSTRACT

Uptake of ^{244}Cm by Bahia grass, clover, corn, soybean, and wheat, as influenced by the addition of lime or lime and chelate to soil, was investigated in a greenhouse experiment. The soil used was from the floodplain of a stream which had received liquid effluents from a nuclear fuel chemical separations facility and contained 0.8 pCi $^{244}Cm/g$. Uptake of Cm by these crops was low with average concentration ratios of 10^{-3}. Lime addition decreased uptake in most cases by a factor of two or more. Uptake response to chelate addition of the limed treatment was not very noticeable. The five crop species showed only slight differences in uptake. Only minimal quantities of ^{244}Cm were incorporated into edible portions. Human consumption of crops grown in this soil would pose little health hazard, based on ICRP standards.

Research on plant assimilation of actinides has been conducted on soils contaminated by weapons testing (Au77, Ny76, Ro76), spiking (Ad75, Ad80a, Ba78, Sc77, Sc80, Th69), or liquid wastes (Ad80b, Ha76, Ha78), but less information is available on assimilation from soils which receive chronic low-level releases of actinides from nuclear fuel chemical separations facilities (Ad80b, Mc80a).

At the Department of Energy's Savannah River Plant (SRP) located near Aiken, South Carolina, two operating nuclear fuel chemical separations facilities exist. These facilities have been in operation for 25 years and release to the environment both low-level aerial and liquid-waste effluents. Each effluent is unique in radionuclide composition due both to the type of release (liquid or aerial) and the facility from which they emanate. Since the 1955 start-up, total Pu releases of 3.6 Ci to the atmosphere, 13.9 Ci to the seepage basins, and 0.4 Ci to the streams have occurred (As77). Most of the releases were prior to 1970, but low-level releases are still occurring. The nature of the spent fuels being processed assures that low-level releases of ^{241}Am and ^{244}Cm have also occurred. Therefore, the soils which have received these inputs are of considerable interest as they contain a suite of isotopes whose chemical form has been influenced by natural-weathering processes for a period of up to 25 years. It is unlikely that these soils could be duplicated by artificial spiking or weathering processes, but represent a real world situation.

For Pu, plant assimilation via root uptake is apparently only a minor source of contamination under field conditions (Ad80b, Da77, Mc76, Mc80a, Pi79). Americium and Cm appear to be more available for uptake in this pathway than Pu (Ad77, Wa72, Pr73, Sc80). Greenhouse studies on ^{241}Am (applied to soil in the nitrate form) indicate that the addition of lime decreased uptake while a chelating agent (DPTA) increased uptake by crop plants (Ad77). If similar soil amendments increase uptake under field conditions, they could cause elevated concentrations of these radionuclides in food products. Furthermore, transuranic elements that have been incorporated into plant tissues may be bound to organic compounds, such as proteins and lipids, which could cause a greater gut transfer (Su80) than the current ICRP values determined using solutions of inorganic compounds (ICRP72). Thus, it is important to examine actinides which may be internally fixed via root uptake. A preliminary soil survey indicated that only Pu and Cm were in sufficient quantities to pursue this type of research. This paper reports the Cm research which has two main objectives:

1) Determine to what extent different crop species vary in their ability to take up Cm, and

2) Determine what influence common agricultural soil amendments have on root uptake of Cm.

To accomplish these objectives, a study was undertaken of plant uptake under greenhouse conditions using soil contaminated by effluents from a nuclear fuel chemical separations facility. This soil

contains a suite of isotopes more closely resembling the chemical forms fostered by real world processes than any soil to which isotopes have been added as spikes.

MATERIALS AND METHODS

A 3 x 4 complete factorial experiment with three soil treatments and four cropping sequences was used. Soil treatments included no amendments, adding dolomitic lime to raise the pH to 6.7, and liming to pH 6.7 plus a combination of zinc and manganese EDTA chelates (ethylene diamine tetraacetic acid) added at a rate equivalent to 3.4 and 1.2 kg/ha, respectively. Each treatment was replicated seven times. Commercial potting soil was used in an additional seven pots per crop to serve as controls to determine whether aerial cross contamination between pots occurred in the greenhouse. The four cropping sequences were: 1) winter wheat-soybean rotation, 2) fallow field-corn rotation, 3) continuous Bahia grass, and 4) continuous white clover. Species used were Triticum aestivum L. var. Coker 68-19, Glycine max (L.) Merrill var. Bragg, Zea mays L. var. Coker 71, Paspalum notatum var. saurae Parodi and Trifolium repens L. All pots of a crop were grouped together with soil treatments randomly located within a crop.

Soil was collected in the early fall of 1976 from the floodplain of a stream receiving primarily liquid effluents from a chemical separations facility. The ^{244}Cm was apparently deposited when flood conditions existed and particulate material settled out on the floodplain (J. E. Pinder, III, pers. com.). Only the upper soil layer (0-15 cm) was collected to ensure as high a concentration of ^{244}Cm as possible. The soil was air dried, passed through a 2-mm mesh sieve and homogenized. Samples were collected from the bulk soil for ^{244}Cm determinations. Soil texture, by the hydrometer method (Bo51), pH, using a 1:1 dilution with deionized water, total carbon, using a Leco Carbon Analyzer, and cation exchange capacity, using sodium acetate (pH 8.2) as the extractant (Ja58) were determined.

Commercial fertilizer (10-10-10), lime, and chelating agents were pre-mixed with small volumes of soil and mixed well with the bulk soil on a per pot basis. Total soil dry weight per pot was 12 kg. Pot design was a modification of that used by Schulz (Sc76) with a fiberglass mat having holes punched in it to allow plant emergence. This mat prevented soil splattering during irrigation and dry particle resuspension. Due to difference in growth form of these species, different numbers of holes in the fiberglass mat were used. Bahia grass and clover had seven holes in the fiberglass mat, wheat and soybean had five and corn had one. Wheat, clover, and Bahia grass were planted in late November 1976. The corn pots were left fallow for the winter and planted the following April. Following wheat harvest, fiberglass mats were replaced and soybeans planted in these pots. New fiberglass mats could not be used following harvest of Bahia grass and clover due to growth form of the plants. All pots were watered as necessary with deionized water. Edible crops were harvested upon maturity and separated into reproductive (grain, bean, ear) and vegetative portions. Forage crops were harvested when appro-

priate. Bahia grass harvests were made in May, June, July, and August. Clover cuttings were made in April, June, and October. These individual samples were combined into one yearly sample per pot. Corn ears from all replication pots were composited. In harvesting, all plants were clipped 5 cm above the upper fiberglass layer to avoid possible soil-particle contamination. Ashed plant (500°C for seven days) and soil samples were sent to LFE Environmental Analysis Laboratories (Richmond, CA) for ^{244}Cm determinations by a total dissolution method (We78). Counting was accomplished using a low-level alpha spectrometry system. Concentration ratios (CR) were computed by dividing the ^{244}Cm concentration of the plant sample by the corresponding ^{244}Cm concentration of the soil.

Analysis of variance was conducted using the Statistical Analysis System (Ba76). Tukey's test was used for comparisons of means within groups (Ki68). There is considerable discussion regarding the best method to treat samples which are below the detection limit. We have chosen to report the values of the detection limit (^{244}Cm = 22.5 fCi/sample) for values below the detection limit. Concentration ratios were calculated taking into account values below the detection limit. The importance of the number of samples which had detectable concentrations can be evaluated since the number of samples used in the computations is indicated in the table.

RESULTS

The actinide contents of the floodplain soil, which primarily received liquid effluents, is dominated by ^{238}Pu, with some ^{244}Cm and very little $^{239,240}Pu$ or ^{241}Am (Table 1). The clay and total carbon content of the floodplain soil is slightly higher than those reported for upland sites (Mc80b). Transuranic elements are known to be fixed by clay minerals and complexed by organic matter which may affect their availability (Sh79).

Of the vegetative portions of corn, wheat, and soybean, 75% of these samples had detectable ^{244}Cm concentrations. Vegetation concentrations resulting from uptake from non-amended soils showed corn > soybean > wheat (Table 2). Lime decreased ^{244}Cm uptake in these three crops, but the only statistically significant differences occurred in corn leaves and stalks. The addition of chelate to the limed soil slightly increased uptake, resulting in means not statistically significant from the limed soil nor equal to the greater uptake observed from the non-amended soil. Corn leaves contained more ^{244}Cm than did the stalks. Concentration ratios showed similar patterns in species and amendment effects (non-amended, CR = 10^{-2} to 10^{-3}; amended, CR = 10^{-3}).

Only 16% of the samples of edible portions of corn, wheat, and soybean contained detectable ^{244}Cm concentrations. Grain, ears, and beans generally had lower concentrations and concentration ratios than the vegetative portions, although the differences were not large.

With 88% of the Bahia grass and clover samples having detectable ^{244}Cm concentrations, the effect of lime became more pronounced with statistically lower uptake of ^{244}Cm for both crops in limed soil. Slightly increased uptake by clover from limed soils was observed when chelate was present. Clover had the greatest uptake rate of all species grown in this experiment.

DISCUSSION

This study was conducted in the greenhouse due to the necessity to shield the plants from global fallout and possible aerial releases from the SRP. These sources can easily obscure uptake and translocation from the roots. Greenhouse results are difficult to extrapolate to field responses because of differences in climate and root growing conditions. However, greenhouse experiments can provide information relative to the effect of environmental factors on Cm phytoavailability which are often difficult to obtain from field experiments. We expect the uptake trends observed in this greenhouse study to follow results in similar root uptake studies in the field. Thus, these results can still effectively evaluate common agricultural amendments that farmers would use to optimize production of the crops involved here. The fact that we used naturally-weathered soil should make the results more valid than comparable spiking experiments, since retention of spiked actinides by most soils does not cease for four to six months (Sh79).

Scientists investigating actinide uptake by plants from soil share the belief that cation exchange capacity of the soil, its content of organic substances, pH, and type of contamination are important factors influencing uptake.

Curium uptake from non-amended soil was greater than for the two amended treatments. The depressing effect of lime addition to soil and the minimal effect of chelate were consistent for all crops. These observations are in agreement with the common knowledge that high pH generally depresses metal uptake and has also been observed to depress uptake of transuranic elements for other crops and soils (Ad77, Ad80a, Ba78, Ho79, Li76, Wa72). The chelate effect was anticipated to be somewhat greater than that observed but our observation was not totally unexpected due to the low ^{244}Cm concentrations. Also, the high C content of the soil (2.69%) indicates that considerable quantities of humic acids probably exist in soil which could mask the effect of the added EDTA by their own natural chelating effect. Thus with soil ^{244}Cm concentrations up to 0.82 pCi/g, uptake by plants was low ($CR \sim 10^{-3}$) and within the range of CR of 10^{-4} to 10^{-3} reported for ^{244}Cm (Ad80a). Garten (Ga80) found similar concentration ranges and CR values for leaves of box elder trees growing on the White Oak Lake floodplain in Tennessee with soil having a median concentration of 0.541 pCi $^{244}Cm/g \cdot soil$.

On a long-term basis, perennial crops (e.g., those regenerated from root stocks and harvested repeatedly) might be expected to take

up more ^{244}Cm than annual crops (e.g., those which are annually replanted as seeds) due to more extensive and established rooting system and greater interactions between Cm particles and long-lived roots. Also, crop species and even plant parts can differ markedly in their transuranic element contents (Ad80a, Sc80). In testing various plant species, legumes were found to accumulate transuranic elements approximately 10 times more than grasses (Sc80). Also, grain or fruit had much lower CR values than the vegetative parts indicating lower translocation to the reproductive parts.

In this study, forage crops (clover and Bahia grass) and vegetative portions of wheat, corn, and soybeans all had similar ^{244}Cm concentrations, although clover generally had the highest. Edible portions did generally have lower ^{244}Cm contents than vegetative portions. Therefore, these data do not support either the hypotheses that perennials, like clover and Bahia grass, take up more ^{244}Cm than the annuals, or that legumes differ from cereal crops. This is probably because of the short duration of the study. Another possible factor affecting the legume hypothesis is the presence of N-fixing bacteria associated with the roots. Clover was inoculated with these bacteria, while soybeans were not. This is the common agricultural practice for the area as it is felt that native soils have sufficient inoculum in the soil for soybeans. What effect this might have on uptake is unknown. In order to examine the long-term uptake response, this experiment is continuing.

Wheat forage, corn silage, soybean meal, clover, and Bahia grass hay are used for livestock feed. Thus, an intermediary step in the food chain to man is included (feeds to livestock to man). Edible portions (wheat grain, corn ears, and soybean beans) showed concentrations which were generally lower than for vegetative portions, but have a shorter food chain to man (direct consumption). Although the gut-absorption coefficient is thought to be higher for ^{244}Cm than Pu (ICRP72), resultant dose from either pathway to man would be quite low.

In "real world" agricultural field situations, the root uptake pathway by crops may not be the most important as illustrated for Pu, where surficial deposition is more important (Mc80a). Surficial deposition can be largely eliminated by food-handling processes, but internally fixed radionuclides cannot be removed by normal food-handling processes, thus this fraction is the most important to the consumer of these foodstuffs. This study has shown the concentration ratio for ^{244}Cm is approximately 10^{-3}. It will vary only slightly due to soil amendments or crop species. Thus, in areas such as ours, where ^{244}Cm soil concentrations are $\leq$ 1 pCi/g (dry weight), crops would assimilate so little ^{244}Cm by root uptake that consumption of the crops grown on this soil would pose little health hazard.

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Table 1. Physical and chemical characteristics and transuranic element concentrations of Floodplain soil used in greenhouse uptake experiment.

Texture	Sand (%)	69
	Silt (%)	11
	Clay (%)	20
CEC (meq/100 g)		8.9
Initial pH		5.2
Final pH		6.7
Total C (%)		2.69
^{238}Pu (pCi/g)		5.28 ± 0.50[1]
$^{239,240}Pu$ (pCi/g)		0.05 ± 0.01
^{241}Am (pCi/g)		0.01 ± 0.00
^{244}Cm (pCi/g)		0.82 ± 0.41

[1] $\bar{x}$ ± 1 S.D., n = 5.

Table 2. Curium-244 concentrations (fCi/g) and concentration ratios (CR) of crop plants.

	SOIL TREATMENT		
	Non-Amended	Lime	Lime & Chelate
Soybean Veg.[4] - Conc.	10.8 ± 5.0[1] (6)[2]	3.8 ± 1.3 (2)	4.5 ± 1.4 (4)
CR	$(1.3 \pm 0.6) \times 10^{-2}$	$(4.7 \pm 1.5) \times 10^{-3}$	$(5.5 \pm 1.7) \times 10^{-3}$
Soybean Bean - Conc.	3.7 ± 0.8 (2)	2.5 ± 0.8 (2)	3.0 ± 1.2 (2)
CR	$(4.5 \pm 1.0) \times 10^{-3}$	$(3.0 \pm 0.9) \times 10^{-3}$	$(3.6 \pm 1.5) \times 10^{-3}$
Wheat Veg.[4] - Conc.	2.9 ± 1.5 (7)	1.3 ± 0.7 (4)	1.5 ± 0.7 (4)
CR	$(3.5 \pm 1.8) \times 10^{-3}$	$(1.6 \pm 0.9) \times 10^{-3}$	$(1.8 \pm 0.9) \times 10^{-3}$
Wheat Grain - Conc.	2.7 ± 0.6 (1)	1.5 ± 0.3 (0)	2.9 ± 1.2 (2)
CR	$(3.3 \pm 0.8) \times 10^{-3}$	$(1.9 \pm 0.4) \times 10^{-3}$	$(3.6 \pm 1.4) \times 10^{-3}$
Corn Stalk - Conc.	7.9 ± 3.0 B[3] (7)	0.9 ± 0.4 A (4)	3.0 ± 3.6 AB (6)
CR	$(9.7 \pm 3.6) \times 10^{-3}$	$(1.2 \pm 0.5) \times 10^{-3}$	$(3.6 \pm 4.3) \times 10^{-3}$
Corn Leaves - Conc.	19.5 ± 9.9 B (7)	2.0 ± 1.1 A (5)	4.4 ± 2.4 A (7)
CR	$(2.4 \pm 1.2) \times 10^{-2}$	$(2.4 \pm 1.4) \times 10^{-3}$	$(5.3 \pm 3.0) \times 10^{-3}$
Corn Ear[5] - Conc.	2.5 (1)	0.8 (1)	0.6 (1)
CR	3.1×10^{-3}	9.5×10^{-4}	7.8×10^{-4}
Bahia grass - Conc.	6.2 ± 1.1 B (7)	0.6 ± 0.3 A (3)	0.7 ± 0.2 A (6)
CR	$(7.5 \pm 1.3) \times 10^{-3}$	$(7.7 \pm 4.2) \times 10^{-4}$	$(8.2 \pm 2.9) \times 10^{-4}$
Clover - Conc.	39.9 ± 4.9 B (7)	2.2 ± 0.5 A (7)	4.5 ± 1.1 A (7)
CR	$(4.9 \pm 0.6) \times 10^{-2}$	$(2.7 \pm 0.7) \times 10^{-3}$	$(5.5 \pm 1.3) \times 10^{-3}$

[1] Mean ± 1 S.D.

[2] Numbers in parentheses indicate number of samples with detectable concentrations (Maximum = 7).

[3] Significant differences exist at 1% level within a plant species. Unlike letter indicates differences between means (Tukey's test).

[4] Vegetation sample primarily composed of stem tissue, with little leaf tissue.

[5] Corn ears from all replication pots were composited into only one sample.

Uptake of Plutonium From Soils Contaminated By A Nuclear Fuel Chemical Separations Facility[1]

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ABSTRACT

A greenhouse experiment investigated factors influencing root uptake of Pu by agricultural crops from soils receiving effluents from a nuclear fuel chemical separations facility. Results indicate that uptake of ^{238}Pu and $^{239,240}Pu$ from soil was very low (concentrations ratio of 10^{-3} or lower). Large differences in Pu soil concentrations did not produce uptake differences indicating that uptake was independent of soil concentrations in this case. Both liming the soil and chelate addition to limed soils produced only very slight differences in uptake of Pu. Clover and soybean took up more Pu than corn, wheat, and bahiagrass. Reproductive parts of these crops had similar concentrations of Pu to their corresponding vegetative parts. Only very minimal quantities (< 10 fCi/g dry weight) of Pu were incorporated into edible portions of crops grown in soil containing 5 pCi Pu/g and would pose little health hazards to man if ingested.

Additional index words: Actinides, transuranics, nuclear technology, concentration ratio

Widespread application of modern nuclear technology depends largely on whether operations are deemed safe for the human population and the environment. A major objection to the use of nuclear technology is the production of hazardous radionuclides including the actinide elements. Release to the environment of uncontrolled amounts of these radionuclides may have serious consequences, because of the toxicity and/or long physical and biological half-lives of the actinide elements (e.g., ^{235}U, ^{238}U, ^{238}Pu, $^{239,240}Pu$, ^{241}Am, and ^{244}Cm). With the national goal of energy independence in a few decades, reprocessing spent nuclear fuel is very important, not only to recycle unused U, but also to separate Pu to produce additional nuclear fuel. Furthermore, at present, reprocessing of spent fuel remains preferable to direct disposal in terms of economics, material conservation, and long-term environmental protection.

Much research on plant assimilation of the actinides has been conducted on soils which have been contaminated by weapons testing (Au et al., 1977; Nyhan et al., 1976; Romney et al., 1976), spiking (Adams et al., 1975; Adriano et al., 1980b; Schulz, 1977), or liquid wastes (Adriano et al., 1980a; Hakonson et al., 1976; Hakonson and Gallegos, 1978). However, only a limited amount of information is available from soils which receive low-level releases of these nuclides from chemical separations (Adriano et al., 1980a; McLeod et al., 1980).

The United States Department of Energy's Savannah River Plant (SRP) located near Aiken, S. C. has two nuclear fuel chemical separations plants (F and H Areas). Since its inception in the early 1950's, monitoring at source points indicated a total of 3.6 Ci of total Pu have been released to the atmosphere, 13.9 Ci to the seepage basins, and 0.4 Ci to the streams. Most of the aerial releases occurred prior to 1970, but low-level releases are continuing. Thus, the industrial operations at SRP provide an excellent research opportunity to study some of the critical pathways of ^{238}Pu and $^{239,240}Pu$ in typical agricultural operations in the southeastern United States.

Recent research on natural vegetation and crops grown in the vicinity of separations plants at SRP indicate that root uptake of Pu is small compared to surface contamination (Adriano et al., 1980a; McLeod et al., 1980; Pinder et al., 1979). Although root uptake is generally small, transuranic elements that have been incorporated into plant tissues may be chemically bound to proteins, lipids and other organic compounds. Sullivan et al. (1980) indicate that gut transfer of these assimilated compounds may be greater than the current ICRP values (ICRP, 1972) which were determined using solutions of inorganic compounds. Thus it is very important to examine the Pu internally fixed via root uptake. This research attempts to answer two major questions:

1) To what extent do crop species differ in their ability to absorb soil Pu?

2) What influence do common soil amendments have on the availability of soil Pu?

To address these questions a study of plant uptake under greenhouse conditions using soils which have received effluents from nuclear fuel chemical separations factilities has been undertaken.

MATERIALS AND METHODS

The experimental design was a 3 x 4 x 3 complete factorial experiment with three soil treatments, four cropping sequences, and three different soils. Soil treatments were no amendment (Control), liming with an amount of dolomitic lime calculated from the basis of a two week incubation to raise the pH to 6.8 (Lime) and liming to pH 6.8 plus a combination of zinc and manganese chelates (ethylene diamine tetraacetic acid - EDTA) added at a rate of 3.4 and 1.2 kg/ha, respectively (Lime and Chelate). The four cropping sequences were: 1) winter wheat-soybean rotation, 2) fallow-corn rotation, 3) continuous bahiagrass, and 4) continuous white clover. Species used were Triticum aestivum L. var. Coker 68-19, Glycine max (L.) Merrill var. Bragg, Zea mays L. var. Coker 71, Paspalum notatum var. saurae Parodi and Trifolium repens L.

Of the three soils used, two were collected from fields which receive aerial effluents from H-Area and where previous research involving agricultural species had been conducted. The third soil was collected from the floodplain of a stream which receives primarily liquid effluents from the same separations area.

Pot design (Fig. 1) was a modification of that used by Schulz et al. (1976) with a fiberglass mat provided with holes to allow plant emergence. The mat prevented soil splattering during irrigation and dry particle resuspension. Due to differences in growth forms of the species, different numbers of holes in the fiberglass mat were used. Bahiagrass and clover had seven holes in the mat, wheat and soybean had five and corn had one. Each treatment was replicated seven times. An additional seven pots per crop, using commercial potting soil media, served as controls to determine whether aerial cross contamination between pots occurred in the greenhouse. All pots for a crop were grouped together in the greenhouse with the different soils and soil treatments randomly distributed within the group.

Soils were collected in early fall of 1976. Only the upper soil profile (0-15 cm) was collected to insure as high a concentration of Pu as possible. They were air dried, passed through a 2 mm mesh sieve and homogenized. Samples were collected from the bulk soil for Pu determinations. For each soil, texture was determined by the hydrometer method (Bouyoucos, 1951). Also pH, using a 1:1 dilution with deionized water, total carbon, using a Leco Carbon Analyzer; and cation exchange capacity, using sodium acetate (pH 8.2) as the extractant (Jackson, 1958), were measured.

Commercial fertilizer (10-10-10), lime, and chelating agents were pre-mixed with small volumes of soil and mixed well with the bulk soil on a per pot basis. Total soil dry weight per pot was 12 kg.

Wheat, clover and bahiagrass were planted in late November 1976. The corn pots were left fallow for the winter and planted the following

April. Following wheat harvest, soybeans were planted in these same pots. Fiberglass mats were replaced following harvest of the wheat crop. New fiberglass mats could not be used following harvest of clover and bahiagrass due to their growth forms. All pots were watered as necessary with deionized water. Edible crops were harvested upon maturity and separated into reproductive (grain, bean, ear) and vegetative portions. Forage crops were harvested when appropriate.

Bahiagrass harvests were made in May, June, July and August. Clover cuttings were made in April, June and October. In harvesting, all plants were clipped 5 cm above the upper fiberglass mat to avoid soil particle contamination. All harvests of the clover and bahiagrass were combined on a per pot basis for Pu analysis.

Ashed plant and soil samples were sent to LFE Environmental Analysis Laboratories in Richmond, CA for Pu determination by the total dissolution method (Wessman et al., 1978). Each sample was counted for 1000 minutes using a low-level alpha spectrometry system. Recovery of the plutonium averaged 78% using these methods. There is considerable discussion regarding the best method to treat samples whose concentrations are below the detection limit. We have chosen to assign the value of the detection limit to those samples (detection limits: ^{238}Pu = 18 fCi/sample; $^{239,240}Pu$ = 13.5 fCi/sample). The importance of the number of samples which actually had detectable concentrations can still be evaluated since the number of samples with detectable concentrations is indicated in each table. Concentration ratios (CR) were computed by dividing the concentration of a particular radionuclide in the plant sample by the corresponding concentration of that radionuclide in the soil. Means and standard deviations of concentrations and concentration ratios were calculated using the Statistical Analysis System (Barr et al., 1976).

RESULTS

Differences exist in the amounts of ^{238}Pu and $^{239,240}Pu$ in the three soils (Table 1). The Floodplain soil, which received liquid effluents, is dominated by ^{238}Pu, with very little $^{239,240}Pu$. Both Field 1 and 2 soils received aerial releases, with Field 2 farther from the release point. Both fields have higher concentrations of $^{239,240}Pu$ than ^{238}Pu. Soil Pu concentrations in Field 2 are one order of magnitude less than in Field 1. The Floodplain soil, with an extremely high $^{238}Pu/^{239,240}Pu$ ratio has a very different isotopic composition than the other two soils. The soils also differ in physical and chemical properties. The Floodplain soil had higher total C content and Field 2 has lower cation exchange capacity. Therefore, the physical and chemical characteristics of these soils may influence Pu uptake by these crops.

Edible Crops - Vegetative Portions:

Analytical difficulties caused by low concentration of radioisotopes were encountered particularly in ^{238}Pu determinations with only

11% of the samples having concentrations above detection limits (Table 2). Cross contamination between pots was also indicated by the fact that 7% of this sample type showed concentrations above the detection limit when grown in non-contaminated soil.

Although the Floodplain soil contained one and two orders of magnitude greater ^{238}Pu concentrations than Field 1 and 2 soils, respectively, there were no apparent differences in plant concentrations of this isotope when grown on different soils. Corn stalks and leaves of corn and wheat had low ^{238}Pu concentrations, while concentrations in soybean vegetation were usually slightly greater. Plutonium-238 concentration ratios appeared to decrease from Field 2 soil to Field 1 soil to Floodplain soil, but the data are incomplete to be conclusive. The data base is also insufficient to draw any conclusions regarding the effects of soil amendments on availability.

Plutonium-239,240 is more abundant than ^{238}Pu in two of the three soils and consequently detection limit problems were lessened. Detectable concentrations were found in approximately 21% of the samples. Cross contamination was again indicated by concentrations above the detection limit in 11% of the plants grown in non-contaminated soil. Thus cross contamination appears to have occurred in this sample type but is not entirely responsible for the plutonium concentration in all of these samples.

Concentrations in the vegetation grown on the three soils were similar (Table 3), although slightly higher concentrations of $^{239,240}Pu$ were observed in the Floodplain and Field 2 soils. Concentration ratios for ^{238}Pu and $^{239,240}Pu$ are an inverse function of the soil concentrations. Thus, with samples near the detection limit, the observed changes in CR could be due exclusively to differences in soil concentration. Soybean foliage exhibited the greatest $^{239,240}Pu$ concentrations, while corn leaves, corn stalks and wheat foliage were similar to each other, but lower than soybeans.

Edible Crops - Reproductive Portions.

Wheat grain and soybean beans had low concentrations of ^{238}Pu and $^{239,240}Pu$ when grown in all three soils (Tables 4 and 5), with soybeans generally having higher concentrations. More samples had detectable ^{238}Pu concentrations when grown in Floodplain soil than in the other two soils. Concentration ratios of ^{238}Pu and $^{239,240}Pu$ for the reproductive portions showed the same patterns as for the vegetative portions, with Field 2 > Field 1 > Floodplain and Field 2 = Floodplain > Field 1, respectively. This again points out the problems of calculating CR values for few samples with minimally detectable concentrations (approximately 15%) and different soil concentrations. Both concentrations and CR were similar between reproductive and vegetative portions of the same crop. Corn ears had no detectable ^{238}Pu or $^{239,240}Pu$. Samples from non-contaminated soils did not have detectable

concentrations of either isotope, indicating aerial cross contamination did not occur in this sample type.

Forage Crops.

The problem of concentration of radioisotopes being below detection limits was reduced with forage crop samples. Approximately 35% of the samples had detectable concentrations. Plants grown in non-contaminated soils showed no detectable plutonium concentration. Again indicating a lack of aerial cross contamination. Concentrations of ^{238}Pu (Table 6) and $^{239,240}Pu$ (Table 7) were very similar to the concentrations in the annual edible crops. Concentration ratios also show similar patterns to that observed in edible crops. Clover occasionally showed higher concentrations of ^{238}Pu and $^{239,240}Pu$ than bahiagrass and are also reflected in higher CR values for this crop.

DISCUSSION

This study was conducted in the greenhouse to shield the plant from global Pu fallout and aerial releases from the separations plants at SRP. These external contaminations often obscure uptake by roots. The validity of extrapolating greenhouse results to evaluate field conditions is still questionable. However, greenhouse experiments can provide information relative to environmental factors affecting Pu bioavailability which are often difficult to obtain from field experiments.

Two groups of crops were evaluated. It was hypothesized that on a long-term basis, perennial crops (i.e., those regenerated from root stocks and harvested repeatedly) would take up greater amounts of Pu than the annual crops (i.e., those which are annually replanted as seeds) due to more extensive rooting system and possible greater interactions between Pu particles and long-lived roots.

In this study, forage crops (clover and bahiagrass) and vegetative portions of wheat, corn, and soybeans all had similar Pu concentrations although clover or soybean generally had the highest. The data does not support the hypothesis that perennials, like clover and bahiagrass take up more Pu than the annuals, possibly because of the short duration of the study period. This experiment is continuing to evaluate long term effects that might be due to the annual or perennial nature of the species studied.

It was also expected that differences in uptake might be caused by one or a combination of the following factors: Pu concentration in soil, crop species, soil chemical or physical properties or soil amendment.

In spite of large differences in Pu concentrations between soils, Pu uptake from all soils appears generally similar. If any trend can be discerned, it is that as the Pu concentration of the soil increased, the Pu CR values tended to decrease. No adequate explanation can be offered at this time although we feel that this trend was caused by the very low and similar Pu concentrations in plant samples from soils which had different Pu concentrations.

High pH caused by lime addition generally depresses metal uptake whereas chelate addition usually increases uptake. These effects have also been observed with the transuranic elements for various crops and soils (Adriano et al., 1977; Adriano et al., 1980b; Ballou et al., 1978; Hoyt and Adriano, 1979; Lipton and Goldin, 1976; Wallace, 1972). Although there are some trends in our data which might support these observations, in our study, the minimally detectable Pu concentrations restrict our ability to discern any effects of soil amendments (lime or lime plus chelate) on Pu uptake by crops.

Crop species and even plant parts can differ markedly in their Pu concentrations (Adriano et. al., 1980b). Leguminous forage crops, such as alfalfa and clover, generally have higher CR values than cereal crops, like barley and wheat. A slight indication that this was true in our study is indicated by the fact the clover or soybean generally had the higher Pu concentrations. In this study, the grain or the fruit had very similar Pu concentrations and CR values to the vegetative parts indicating no difference in plant parts.

Due to problems of plant concentrations for one or both isotopes being at or below detectable limits in many cases, very few samples could be used to compare the relative uptakes of the two radionuclides. The ratio of radionuclide concentrations was calculated only in instances where this ratio could be developed for two or more samples in at least two soil treatments. Since the isotopic ratio of ^{238}Pu to $^{239,240}Pu$ was greater in clover samples than the same ratio for Field 1 soil, ^{238}Pu appears to be preferentially absorbed (Table 8). The opposite trend was observed for clover grown in Floodplain soils. Also soil treatments affected this ratio in opposite manners in these two soils. Again no adequate explanations can be offered at this time due to insufficient data. However, these discrepancies may be due to extreme differences in soil Pu compositions and/or different-aged Pu particles in various soils, in addition to low detectabilities.

Although soil Pu concentrations ranged up to 5.33 pCi/g, uptake by plants was low (CR values generally less than 10^{-3}). This is within the range of CR values of 10^{-7} to 10^{-3} for $^{239,240}Pu$ for numerous crops and lower than the CR values for the other transuranics of 10^{-4} to 10^{-1} for ^{241}Am, 10^{-4} to 10^{-3} for ^{244}Cm and 10^{-2} to 10^{-1} for ^{237}Np (Adriano et al. 1980b). Wheat forage, corn silage, soybean meal, clover and bahiagrass are used for livestock feed, thus include an intermediate step in the food chain to man (feeds to livestock to man). Edible portions (wheat grain, corn ears, and soybean beans) had concentrations which were generally similar to vegetative portions, but have a shorter

food chain to man (direct consumption). This dose from consumption of edible foodstuffs grown in these soils would be very low in comparison to the 120 mrem annual background radiation dose received in the Carolinas. Resultant dose from forage crops would be considerally lower due to the intermediary step in the food chain.

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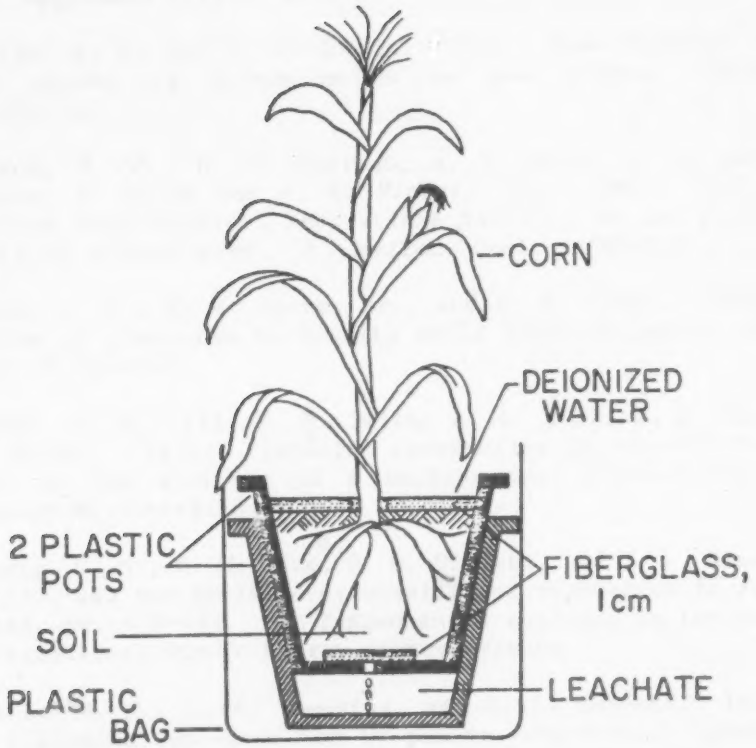

Fig. 1. Pot design.

Table 1. Physical, chemical and plutonium characterization of soils used in greenhouse root uptake experiment.

Soil	Floodplain	Field 1	Field 2
Texture			
Sand, %	69	70	77
Silt, %	11	7	8
Clay, %	20	23	16
CEC, meq/100 g	8.9	8.5	5.3
Initial pH	5.2	6.4	4.9
Total C, %	2.69	1.12	1.01
^{238}Pu, pCi/g	5.28 ± 0.50[1]	0.71 ± 0.09	0.05 ± 0.006
$^{239,240}Pu$, pCi/g	0.05 ± 0.01	2.28 ± 0.41	0.13 ± 0.02
$^{238}Pu/^{239,240}Pu$	105.60	0.31	0.38

[1] $\bar{x}$ ± 1 S.D., n = 5.

Table 2. Plutonium-238 concentrations (fCi/g·dry weight) of vegetative portions of annual crops.

SOIL TREATMENT	Control	Lime	Lime & Chelate
		Floodplain Soil	
Soybean[3] - Conc.	3.6 ± 0.7[1] (0)[2]	2.7 ± 0.5 (1)	4.6 ± 4.7 (3)
CR	$(6.8 \pm 1.3) \times 10^{-4}$	$(5.1 \pm 1.0) \times 10^{-4}$	$(8.7 \pm 8.9) \times 10^{-4}$
Wheat[4] - Conc.	1.3 ± 1.1 (2)	1.0 ± 0.8 (2)	1.1 ± 1.1 (1)
CR	$(2.5 \pm 2.1) \times 10^{-4}$	$(1.8 \pm 1.6) \times 10^{-4}$	$(2.0 \pm 2.1) \times 10^{-4}$
Corn Stalk[5] - Conc.	1.1 ± 0.7 (0)	0.6 ± 0.2 (0)	0.8 ± 0.8 (0)
CR	$(2.1 \pm 1.2) \times 10^{-4}$	$(1.1 \pm 0.4) \times 10^{-4}$	$(1.5 \pm 1.5) \times 10^{-4}$
Corn Leaves[6] - Conc.	0.9 ± 0.3 (0)	0.8 ± 0.2 (1)	0.9 ± 0.3 (1)
CR	$(1.8 \pm 0.7) \times 10^{-4}$	$(1.5 \pm 0.3) \times 10^{-4}$	$(1.7 \pm 0.5) \times 10^{-4}$
		Field 1 Soil	
Soybean[3] - Conc.	2.1 ± 0.5 (1)	2.9 ± 1.2 (1)	2.7 ± 1.0 (1)
CR	$(3.0 \pm 0.7) \times 10^{-3}$	$(4.1 \pm 1.7) \times 10^{-3}$	$(3.8 \pm 1.5) \times 10^{-3}$
Wheat[4] - Conc.	0.5 ± 0.0 (0)	0.5 ± 0.0 (0)	0.5 ± 0.1 (1)
CR	$(6.8 \pm 0.5) \times 10^{-4}$	$(6.8 \pm 0.6) \times 10^{-4}$	$(6.4 \pm 1.5) \times 10^{-4}$
Corn Stalk[5] - Conc.	0.5 ± 0.3 (0)	0.6 ± 0.4 (0)	0.4 ± 0.2 (0)
CR	$(6.8 \pm 4.9) \times 10^{-4}$	$(8.8 \pm 5.4) \times 10^{-4}$	$(5.9 \pm 3.1) \times 10^{-4}$
Corn Leaves[6] - Conc.	0.5 ± 0.1 (1)	0.6 ± 0.2 (1)	0.5 ± 0.1 (1)
CR	$(7.5 \pm 1.8) \times 10^{-4}$	$(8.9 \pm 2.8) \times 10^{-4}$	$(7.0 \pm 1.4) \times 10^{-4}$
		Field 2 Soil	
Soybean[3] - Conc.	7.9 ± 3.1 (1)	2.4 ± 0.7 (0)	1.8 ± 0.6 (0)
CR	$(1.6 \pm 0.6) \times 10^{-1}$	$(4.8 \pm 1.5) \times 10^{-2}$	$(3.7 \pm 1.2) \times 10^{-2}$
Wheat[4] - Conc.	0.7 ± 0.2 (1)	0.5 ± 0.1 (1)	0.5 ± 0.0 (1)
CR	$(1.4 \pm 0.4) \times 10^{-2}$	$(9.2 \pm 2.1) \times 10^{-3}$	$(9.3 \pm 0.5) \times 10^{-3}$
Corn Stalk[5] - Conc.	1.8 ± 1.8 (0)	0.5 ± 0.3 (0)	1.0 ± 0.6 (1)
CR	$(3.6 \pm 3.7) \times 10^{-2}$	$(9.4 \pm 5.9) \times 10^{-3}$	$(2.0 \pm 1.3) \times 10^{-2}$
Corn Leaves[6] - Conc.	0.9 ± 0.2 (1)	0.8 ± 0.7 (2)	0.9 ± 0.4 (0)
CR	$(1.9 \pm 0.5) \times 10^{-2}$	$(1.7 \pm 1.4) \times 10^{-2}$	$(1.7 \pm 0.9) \times 10^{-2}$

[1] $\bar{x} \pm 1$ S.D. Calculations used the value of the detection limit for those samples whose plutonium content was less than the detection limit.

[2] Number in parenthesis indicates number of samples with detectable concentrations. Maximum = 7.

[3] Sample weights ranged from 1.43 to 17.76g, $\bar{x}$ = 7.24.

[4] Sample weights ranged from 18.73 to 43.42g, $\bar{x}$ = 33.49.

[5] Sample weights ranged from 3.14 to 74.07g, $\bar{x}$ = 35.39.

[6] Sample weights ranged from 10.90 to 51.90g, $\bar{x}$ = 28.67.

Table 3. Plutonium-239,240 concentrations (fCi/g·dry weight) of vegetative portions of annual crops. Sample weights are given in Table 2.

SOIL TREATMENT	Control	Lime	Lime & Chelate
		Floodplain Soil	
Soybean - Conc.	2.7 ± 0.5[1] (1)[2]	1.9 ± 0.5 (1)	1.9 ± 0.4 (3)
CR	$(5.4 \pm 1.0) \times 10^{-2}$	$(3.9 \pm 0.9) \times 10^{-2}$	$(3.7 \pm 0.8) \times 10^{-2}$
Wheat - Conc.	0.7 ± 0.3 (3)	0.4 ± 0.1 (2)	0.5 ± 0.1 (1)
CR	$(1.3 \pm 0.6) \times 10^{-2}$	$(8.8 \pm 1.8) \times 10^{-3}$	$(9.6 \pm 1.3) \times 10^{-3}$
Corn Stalk - Conc.	0.8 ± 0.5 (0)	0.4 ± 0.2 (0)	0.6 ± 0.6 (0)
CR	$(1.7 \pm 1.0) \times 10^{-2}$	$(8.8 \pm 3.2) \times 10^{-3}$	$(1.2 \pm 1.2) \times 10^{-2}$
Corn Leaves - Conc.	0.7 ± 0.3 (0)	0.6 ± 0.2 (1)	0.8 ± 0.5 (2)
CR	$(1.4 \pm 0.5) \times 10^{-2}$	$(1.2 \pm 0.3) \times 10^{-2}$	$(1.6 \pm 0.9) \times 10^{-2}$
		Field 1 Soil	
Soybean - Conc.	1.6 ± 0.4 (1)	2.3 ± 0.9 (1)	2.5 ± 1.9 (3)
CR	$(7.2 \pm 1.6) \times 10^{-4}$	$(9.9 \pm 3.8) \times 10^{-4}$	$(1.1 \pm 0.8) \times 10^{-3}$
Wheat - Conc.	0.4 ± 0.1 (2)	0.5 ± 0.3 (2)	0.6 ± 0.5 (3)
CR	$(1.8 \pm 0.5) \times 10^{-4}$	$(2.1 \pm 1.1) \times 10^{-4}$	$(2.4 \pm 2.0) \times 10^{-4}$
Corn Stalk - Conc.	0.4 ± 0.3 (0)	0.5 ± 0.3 (0)	0.3 ± 0.2 (0)
CR	$(1.6 \pm 1.1) \times 10^{-4}$	$(2.0 \pm 1.3) \times 10^{-4}$	$(1.4 \pm 0.7) \times 10^{-4}$
Corn Leaves - Conc.	0.5 ± 0.1 (4)	0.5 ± 0.2 (0)	0.5 ± 0.2 (4)
CR	$(2.1 \pm 0.5) \times 10^{-4}$	$(2.1 \pm 0.7) \times 10^{-4}$	$(2.1 \pm 0.9) \times 10^{-4}$
		Field 2 Soil	
Soybean - Conc.	6.1 ± 2.3 (0)	4.5 ± 6.5 (2)	1.9 ± 1.8 (1)
CR	$(4.7 \pm 1.8) \times 10^{-2}$	$(3.5 \pm 5.0) \times 10^{-2}$	$(1.5 \pm 1.4) \times 10^{-2}$
Wheat - Conc.	0.5 ± 0.1 (2)	0.5 ± 0.3 (5)	0.5 ± 0.2 (3)
CR	$(4.1 \pm 1.0) \times 10^{-3}$	$(3.6 \pm 2.5) \times 10^{-3}$	$(3.6 \pm 1.3) \times 10^{-3}$
Corn Stalk - Conc.	1.4 ± 1.4 (1)	0.4 ± 0.2 (0)	0.7 ± 0.4 (1)
CR	$(1.0 \pm 1.1) \times 10^{-2}$	$(2.7 \pm 1.7) \times 10^{-3}$	$(5.5 \pm 3.4) \times 10^{-3}$
Corn Leaves - Conc.	0.8 ± 0.3 (1)	0.4 ± 0.2 (1)	0.7 ± 0.3 (2)
CR	$(6.1 \pm 2.6) \times 10^{-3}$	$(3.4 \pm 1.3) \times 10^{-3}$	$(5.7 \pm 2.7) \times 10^{-3}$

[1] $\bar{x} \pm 1$ S.D. Calculations used the value of the detection limit for those samples whose plutonium content was less than the detection limit.

[2] Number in parenthesis indicates number of samples with detectable concentrations. Maximum = 7.

Table 4. Plutonium-238 concentrations (fCi/g·dry weight) of edible portions of annual crops.

SOIL TREATMENT		Control	Lime	Lime & Chelate
			Floodplain Soil	
Soybean Bean[3]	Conc.	2.7 ± 0.6[1] (2)[2]	1.6 ± 0.2 (2)	1.8 ± 1.0 (2)
	CR	$(5.2 \pm 1.1) \times 10^{-4}$	$(3.1 \pm 0.4) \times 10^{-4}$	$(3.5 \pm 1.9) \times 10^{-4}$
Wheat Grain[4]	Conc.	2.0 ± 0.7 (1)	1.2 ± 0.3 (1)	1.5 ± 0.5 (2)
	CR	$(3.7 \pm 1.3) \times 10^{-4}$	$(2.3 \pm 0.6) \times 10^{-4}$	$(2.8 \pm 0.9) \times 10^{-4}$
			Field 1 Soil	
Soybean Bean[3]	Conc.	1.7 ± 0.5 (0)	1.6 ± 0.6 (1)	1.7 ± 0.9 (1)
	CR	$(2.4 \pm 0.7) \times 10^{-3}$	$(2.2 \pm 0.8) \times 10^{-3}$	$(2.4 \pm 1.3) \times 10^{-3}$
Wheat Grain[4]	Conc.	1.3 ± 0.8 (1)	1.0 ± 0.1 (0)	1.2 ± 0.8 (0)
	CR	$(1.8 \pm 1.1) \times 10^{-3}$	$(1.4 \pm 0.2) \times 10^{-3}$	$(1.6 \pm 1.2) \times 10^{-3}$
			Field 2 Soil	
Soybean Bean[3]	Conc.	12.9 ± 15.9 (0)	1.6 ± 0.4 (0)	1.5 ± 0.6 (2)
	CR	$(2.6 \pm 3.2) \times 10^{-1}$	$(3.2 \pm 0.8) \times 10^{-2}$	$(3.0 \pm 1.3) \times 10^{-2}$
Wheat Grain[4]	Conc.	1.7 ± 0.9 (2)	1.4 ± 0.6 (3)	0.9 ± 0.1 (0)
	CR	$(3.5 \pm 1.7) \times 10^{-2}$	$(2.8 \pm 1.3) \times 10^{-2}$	$(1.9 \pm 0.3) \times 10^{-2}$

[1] $\bar{X} \pm 1$ S.D. Calculations used the value of the detection limit for those samples whose plutonium content was less than the detection limit.

[2] Number in parenthesis indicates number of samples with detectable concentrations. Maximum = 7.

[3] Sample weights ranged from 0.37 to 19.67g, $\bar{x} = 10.09$.

[4] Sample weights ranged from 4.84 to 24.88g, $\bar{x} = 15.33$.

Table 5. Plutonium-239,240 concentrations (fCi/g·dry weight) of edible portions of annual crops. Sample weights are given in Table 4.

SOIL TREATMENT	Control	Lime	Lime & Chelate
		Floodplain Soil	
Soybean Bean - Conc.	2.0 ± 0.2[1] (0)[2]	1.5 ± 0.4 (1)	1.5 + 0.7 (0)
CR	$(3.9 \pm 0.4) \times 10^{-2}$	$(2.9 \pm 0.8) \times 10^{-2}$	$(3.0 \pm 1.3) \times 10^{-2}$
Wheat Grain - Conc.	1.5 ± 0.5 (1)	0.9 ± 0.2 (2)	1.3 ± 0.7 (2)
CR	$(2.9 \pm 1.0) \times 10^{-2}$	$(1.9 \pm 0.3) \times 10^{-2}$	$(2.6 \pm 1.4) \times 10^{-2}$
		Field 1 Soil	
Soybean Bean - Conc.	1.3 ± 0.4 (0)	1.1 ± 0.3 (1)	1.4 ± 0.7 (0)
CR	$(5.6 \pm 1.6) \times 10^{-4}$	$(4.9 \pm 1.2) \times 10^{-4}$	$(5.9 \pm 3.0) \times 10^{-4}$
Wheat Grain - Conc.	1.0 ± 0.6 (3)	0.8 ± 0.1 (0)	0.9 ± 0.6 (0)
CR	$(4.2 \pm 2.8) \times 10^{-4}$	$(3.3 \pm 0.5) \times 10^{-4}$	$(3.9 \pm 2.8) \times 10^{-4}$
		Field 2 Soil	
Soybean Bean - Conc.	9.7 ± 11.9 (0)	1.2 ± 0.3 (0)	1.1 ± 0.5 (0)
CR	$(7.4 \pm 9.1) \times 10^{-2}$	$(9.2 \pm 2.2) \times 10^{-3}$	$(8.5 \pm 3.7) \times 10^{-3}$
Wheat Grain - Conc.	1.3 ± 0.6 (3)	1.3 ± 0.9 (2)	0.9 ± 0.6 (2)
CR	$(1.0 \pm 0.5) \times 10^{-2}$	$(9.9 \pm 6.9) \times 10^{-3}$	$(6.8 \pm 4.3) \times 10^{-3}$

[1] $\bar{x} \pm 1$ S.D. Calculations used the value of the detection limit for those samples whose plutonium content was less than the detection limit.

[2] Number in parenthesis indicates number of samples with detectable concentrations. Maximum = 7.

Table 6. Plutonium-238 concentrations (fCi/g·dry weight) of perennial forage crops.

SOIL TREATMENT	Control	Lime	Lime & Chelate
		Floodplain Soil	
Bahia[3] - Conc.	0.3 ± 0.1[1] (5)[2]	0.3 ± 0.0 (0)	0.3 ± 0.2 (3)
CR	(6.6 ± 2.4) x 10^{-5}	(6.6 ± 0.9) x 10^{-5}	(6.2 ± 3.1) x 10^{-5}
Clover[4] - Conc.	2.2 ± 1.1 (6)	0.4 ± 0.0 (2)	0.8 ± 0.3 (6)
CR	(4.2 ± 2.0) x 10^{-4}	(8.2 ± 0.8) x 10^{-5}	(1.5 ± 0.7) x 10^{-4}
		Field 1 Soil	
Bahia[3] - Conc.	0.3 ± 0.0 (0)	0.3 ± 0.1 (3)	0.3 ± 0.0 (2)
	(4.0 ± 0.5) x 10^{-4}	(4.0 ± 1.4) x 10^{-4}	(3.7 ± 0.7) x 10^{-4}
Clover[4] - Conc.	0.4 ± 0.2 (2)	0.4 ± 0.1 (4)	0.4 ± 0.1 (2)
CR	(6.1 ± 2.2) x 10^{-4}	(5.0 ± 1.4) x 10^{-4}	(5.0 ± 1.7) x 10^{-4}
		Field 2 Soil	
Bahia[3] - Conc.	0.3 ± 0.0 (3)	0.2 ± 0.1 (2)	0.3 ± 0.1 (1)
CR	(5.6 ± 0.8) x 10^{-3}	(5.0 ± 1.7) x 10^{-3}	(5.7 ± 1.1) x 10^{-3}
Clover[4] - Conc.	3.3 ± 2.4 (1)	0.5 ± 0.1 (2)	0.5 ± 0.1 (0)
CR	(6.7 ± 4.8) x 10^{-2}	(9.9 ± 2.7) x 10^{-3}	(1.0 ± 0.2) x 10^{-2}

[1] $\bar{x}$ ± 1 S.D. Calculations used the value of the detection limit for those samples whose plutonium content was less than the detection limit.

[2] Number in parenthesis indicates number of samples with detectable concentrations. Maximum = 7.

[3] Sample weights ranged from 35.75 to 78.61g, $\bar{x}$ = 58.77.

[4] Sample weights ranged from 2.29 to 65.50g, $\bar{x}$ = 37.70.

Table 7. Plutonium-239,240 concentrations (fCi/g·dry weight) of perennial forage crops. Sample weights are given in Table 6.

SOIL TREATMENT	Control	Lime	Lime & Chelate
		Floodplain Soil	
Bahia - Conc.	0.2 ± 0.1[1] (4)[2]	0.2 ± 0.0 (2)	0.3 ± 0.1 (2)
CR	$(4.4 \pm 1.2) \times 10^{-3}$	$(4.9 \pm 0.4) \times 10^{-3}$	$(6.1 \pm 2.3) \times 10^{-3}$
Clover - Conc.	1.0 ± 1.5 (1)	0.3 ± 0.1 (3)	0.3 ± 0.1 (3)
CR	$(2.0 \pm 2.9) \times 10^{-2}$	$(5.8 \pm 1.2) \times 10^{-3}$	$(5.6 \pm 2.3) \times 10^{-3}$
		Field 1 Soil	
Bahia - Conc.	0.2 ± 0.0 (2)	0.2 ± 0.1 (3)	0.2 ± 0.0 (2)
CR	$(8.9 \pm 1.3) \times 10^{-5}$	$(1.1 \pm 0.3) \times 10^{-4}$	$(8.3 \pm 1.6) \times 10^{-5}$
Clover - Conc.	0.3 ± 0.1 (5)	0.3 ± 0.1 (5)	0.4 ± 0.2 (3)
CR	$(1.4 \pm 0.4) \times 10^{-4}$	$(1.2 \pm 0.5) \times 10^{-4}$	$(1.7 \pm 0.7) \times 10^{-4}$
		Field 2 Soil	
Bahia - Conc.	0.3 ± 0.2 (3)	0.2 ± 0.0 (2)	0.2 ± 0.1 (2)
CR	$(2.3 \pm 1.3) \times 10^{-3}$	$(1.6 \pm 0.3) \times 10^{-3}$	$(1.7 \pm 0.4) \times 10^{-3}$
Clover - Conc.	2.5 ± 1.8 (0)	0.3 ± 0.1 (3)	0.4 ± 0.1 (1)
CR	$(1.9 \pm 1.4) \times 10^{-2}$	$(2.6 \pm 0.5) \times 10^{-3}$	$(3.0 \pm 0.7) \times 10^{-3}$

[1] $\bar{x} \pm 1$ S.D. Calculations used the value of the detection limit for those samples whose plutonium content was less than detection limit.

[2] Number in parenthesis indicates number of samples with detectable concentrations. Maximum = 7.

Table 8. Ratios of plant and soil concentrations of ^{238}Pu and $^{239,240}Pu$.

SOIL TREATMENT	Plant samples			Soil samples
	Control	Lime	Lime & Chelate	
		$^{238}Pu/^{239,240}Pu$		
Field 1 - Clover	2.08 ± 0.82[1] (2)[2]	1.60 ± 1.22 (3)	0.43 ± 0.33 (2)	0.31
Floodplain - Clover	0.33 (1)	1.67 ± 0.47 (2)	7.00 ± 3.46 (3)	105.60

[1] $\bar{x} \pm 1$ S. D.

[2] Number in parenthesis indicates the number of samples with detectable concentration of both radionuclides.

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16. ABSTRACT (200 words or less)

Our studies investigating the environmental behavior of radionuclides released into agricultural systems by a nuclear fuel chemical separations facility have advanced considerable within the past year. Manuscripts reporting uptake of Pu and Cm by agricultural plants have shown that Cm was taken up in greater quantities than Pu. Lime addition to soil depresses uptake of Cm while chelate addition to limed soil increases uptake of Cm. No conclusive effects of soil amendments were observed with regard to Pu uptake. Cesium uptake by agricultural plants differs between species, plant part and year of harvest, with an indication that cesium may be increasingly taken up with the passage of time in some instances. Soil amendments affect 137Cs uptake similarly as that reported for 244Cm Rice varities do not differ in their Pu or Cm uptake, nor does uptake by rice in flooded soils differ from those uptake rates reported for dry-land agricultural species. Curium and 239,240Pu appear to be equally available to rice and more available than 238Pu. Broadleaf crops appear to exhibit Pu concentrations dependent on their shape and form, indicating the importance of physical processes in this field experiment. Because of crop failures caused by drought conditions, there has been little progress on our U-field studies.

17. KEY WORDS AND DOCUMENT ANALYSIS

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17b. IDENTIFIERS/OPEN-ENDED TERMS

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